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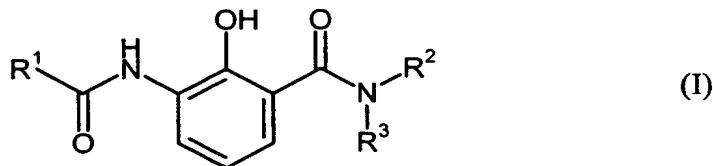
Aminosalicylamides

JC13 Rec'd PCT/PTO 12 FEB 2002

The invention relates to known and novel acylaminosalicylamides, to a plurality of processes for their preparation and to their use for controlling plant-damaging organisms, and also to novel intermediates and processes for their preparation.

Certain aminosalicylamides, and their fungicidal action, are already known (compare, for example, WO 97-08135, WO 98-41513 or WO 99-27783). However, the activity of these prior-art compounds is, in particular at low application rates and concentrations, not entirely satisfactory in all areas of use.

It has now been found that the acylaminosalicylamides of the general formula (I),

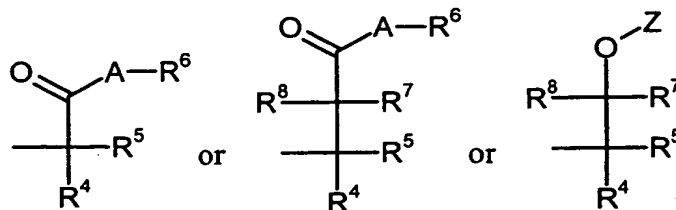


in which

R<sup>1</sup> represents hydrogen or alkyl,

R<sup>2</sup> represents hydrogen or alkyl, or

R<sup>3</sup> represents a grouping



in which

A represents oxygen, sulphur or -(N-R<sup>9</sup>)- in which

$R^9$  represents hydrogen or alkyl or together with  $R^6$  and the nitrogen atom to which they are attached forms an optionally substituted heterocyclic ring,

5  $R^4$  represents hydrogen, optionally substituted alkyl or optionally substituted aryl or

$R^2$  and  $R^4$  together with the atoms to which they are attached form a heterocyclic ring,

10

$R^5$  represents hydrogen or alkyl or

$R^4$  and  $R^5$  together with the carbon atom to which they are attached form a carbocyclic ring,

15

$R^6$  represents hydrogen or in each case optionally substituted alkyl, cycloalkyl, aryl or heterocyclyl,

$R^7$  represents hydrogen or alkyl,

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$R^8$  represents hydrogen or alkyl and

$Z$  represents hydrogen or in each case optionally substituted alkyl, alkylcarbonyl, cycloalkyl, cycloalkylcarbonyl, aryl, arylcarbonyl, heterocyclyl or heterocyclylcarbonyl,

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are suitable for controlling organisms causing damage to plants and industrial materials. These organisms are to be understood as meaning, in particular, microorganisms.

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In the definitions, the hydrocarbon chains, such as alkyl, alkylene, alkenyl or alkynyl, are in each case straight-chain or branched, including in combination with heteroatoms, such as in alkoxy, alkylthio or alkylamino. Unless stated otherwise, preference is given to hydrocarbon chains having 1 to 6 carbon atoms.

5

Halogen generally represents fluorine, chlorine, bromine or iodine, preferably fluorine, chlorine or bromine, in particular fluorine or chlorine.

10

Aryl represents aromatic, mono- or polycyclic hydrocarbon rings, such as, for example, phenyl, naphthyl, anthranyl, phenanthryl, preferably phenyl or naphthyl, in particular phenyl.

15

Heterocyclyl represents saturated or unsaturated, and also aromatic, cyclic compounds having up to eight ring members in which at least one ring member is a heteroatom, i.e. an atom different from carbon. If the ring contains a plurality of heteroatoms, these can be identical or different. Preferred heteroatoms are oxygen, nitrogen and sulphur. If the ring contains a plurality of oxygen atoms, these are not directly adjacent. If appropriate, the cyclic compounds form, together with further carbocyclic or heterocyclic, fused-on or bridged rings, a polycyclic ring system. Preference is given to mono- or bicyclic ring systems, in particular to mono- or bicyclic aromatic ring systems.

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25

Cycloalkyl represents saturated carbocyclic cyclic compounds which form, if appropriate, a polycyclic ring system with other carbocyclic, fused-on or bridged rings.

30

Cycloalkenyl represents carbocyclic cyclic compounds which contain at least one double bond and which form, if appropriate, a polycyclic ring system with other carbocyclic, fused-on or bridged rings.

Halogenalkoxy represents partially or fully halogenated alkyl. In the case of polyhalogenated halogenoalkoxy, the halogen atoms can be identical or different.

Preferred halogen atoms are fluorine or chlorine, in particular fluorine. If the halogenoalkoxy carries further substituents, the maximum possible number of halogen atoms is reduced to the different free valencies. Unless stated otherwise, preference is given to hydrocarbon chains having 1 to 6 carbon atoms.

5

Halogenalkyl represents partially or fully halogenated alkyl. In the case of polyhalogenated halogenoalkyl, the halogen atoms can be identical or different. Preferred halogen atoms are fluorine or chlorine, in particular fluorine. If the halogenoalkyl carries further substituents, the maximum possible number of halogen atoms is reduced to the different free valencies. Unless stated otherwise, preference is given to hydrocarbon chains having 1 to 6 carbon atoms.

10

If appropriate, the compounds of the formula (I) are present as mixtures of various possible isomeric forms, in particular of stereoisomers such as, for example, E and Z, threo and erythro, and also optical isomers. What is claimed is both the use of E and the Z isomers, and also the threo and erythro and the optical isomers, and any mixtures of these isomers.

15

Preference is given to using compounds of the formula (I) in which

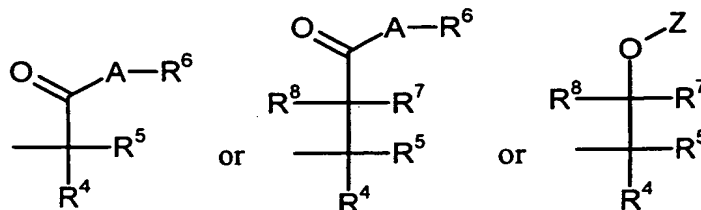
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$R^1$  represents hydrogen or methyl,

$R^2$  represents hydrogen or  $C_1$ - $C_4$ -alkyl and

25

$R^3$  represents a grouping



or

or

in which

$A$  represents oxygen, sulphur or  $-(N-R^9)-$  in which

$R^9$  represents hydrogen or alkyl having 1 to 4 carbon atoms or together with  $R^6$  and the nitrogen atom to which they are attached forms an optionally  $C_1$ - $C_4$ -alkyl-substituted heterocyclic ring having 3 to 7 ring members,

$R^4$  represents hydrogen or alkyl which is optionally substituted by alkoxy, alkylthio, alkoxycarbonyl or alkylcarbonyloxy having in each case 1 to 6 carbon atoms in the alkyl moiety or by arylcarbonyloxy which is optionally substituted in the aryl moiety by hydroxyl, formyloxy, or represents aryl, heterocyclyl, arylalkyl or heterocyclylalkyl having in each case 1 to 6 carbon atoms in the alkyl moiety and being in each case optionally substituted in the aryl moiety or heterocyclyl moiety, or

$R^2$  and  $R^4$  together with the atoms to which they are attached form a heterocyclic ring having 3 to 6 ring members,

$R^5$  represents hydrogen or  $C_1$ - $C_4$ -alkyl or

$R^4$  and  $R^5$  together with the carbon atom to which they are attached form a carbocyclic ring having 3 to 6 ring members,

$R^6$  represents hydrogen or  $C_1$ - $C_{12}$ -alkyl, optionally  $C_1$ - $C_4$ -alkyl-substituted  $C_3$ - $C_7$ -cycloalkyl, or represents aryl, arylalkyl having 1 to 6 carbon atoms in the alkyl moiety, heterocyclyl or heterocyclylalkyl having 1 to 6 carbon atoms in the alkyl moiety, each of which is optionally substituted in the aryl or heterocyclyl moiety,

$R^7$  represents hydrogen or  $C_1$ - $C_4$ -alkyl,

R<sup>8</sup> represents hydrogen or C<sub>1</sub>-C<sub>4</sub>-alkyl and

Z represents hydrogen or C<sub>1</sub>-C<sub>12</sub>-alkyl or alkylcarbonyl, optionally C<sub>1</sub>-C<sub>4</sub>-alkyl-substituted C<sub>3</sub>-C<sub>7</sub>-cycloalkyl or cycloalkylcarbonyl, represents aryl, arylcarbonyl, arylalkyl, arylalkylcarbonyl having 1 to 6 carbon atoms in the alkyl moiety, heterocyclyl, heterocyclylcarbonyl, heterocyclylalkyl or heterocyclylalkylcarbonyl having 1 to 6 carbon atoms in the alkyl moiety, each of which is optionally substituted in the aryl or heterocyclyl moiety.

Preferred substituents for aryl or arylalkyl are given in the list below:

halogen, cyano, amino, hydroxyl, formyl, carboxyl, carbamoyl, thiocarbamoyl;

in each case straight-chain or branched alkyl, alkoxy, alkylthio, alkylsulphinyl or alkylsulphonyl having in each case 1 to 6 carbon atoms;

in each case straight-chain or branched alkenyl or alkenyloxy having in each case 2 to 6 carbon atoms;

in each case straight-chain or branched halogenoalkyl, halogenoalkoxy, halogenoalkylthio, halogenoalkylsulphinyl or halogenoalkylsulphonyl having in each case 1 to 6 carbon atoms and 1 to 13 identical or different halogen atoms;

in each case straight-chain or branched halogenoalkenyl or halogenoalkenyloxy having in each case 2 to 6 carbon atoms and 1 to 13 identical or different halogen atoms;

in each case straight-chain or branched alkylamino, dialkylamino, alkylcarbonyl, alkylcarbonyloxy, alkoxycarbonyl, alkylsulphonyloxy, hydroxyiminoalkyl or

alkoxyiminoalkyl having in each case 1 to 6 carbon atoms in the individual alkyl moieties;

in each case doubly attached alkylene or dioxyalkylene having in each case 1 to 6 carbon atoms and being in each case optionally mono- or polysubstituted by identical or different substituents selected from the group consisting of halogen, straight-chain or branched alkyl having 1 to 4 carbon atoms and straight-chain or branched halogenoalkyl having 1 to 4 carbon atoms and 1 to 9 identical or different halogen atoms; and

cycloalkyl having 3 to 6 carbon atoms, aryl and aryloxy.

Preferred substituents for heterocyclyl or heterocyclalkyl are given in the list below:

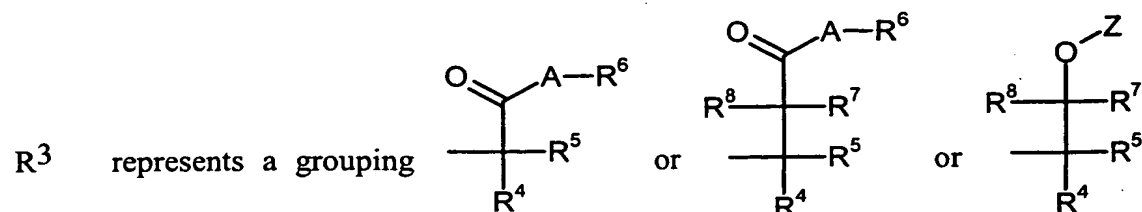
halogen, amino, hydroxyl, oxo,

alkyl, alkoxy, alkylthio, alkylamino, dialkylamino having in each case 1 to 6 carbon atoms in the individual alkyl moieties.

Particular preference is given to using compounds of the formula (I), in which

$R^1$  represents hydrogen or methyl,

$R^2$  represents hydrogen, methyl, ethyl, n- or i-propyl, n-, i-, s- or t-butyl and



in which

A represents oxygen, sulphur or  $-(N-R^9)-$  in which

5  $R^9$  represents hydrogen or methyl, ethyl, n- or i-propyl, n-, i-, s- or t-butyl or together with  $R^6$  and the nitrogen atom to which they are attached represents optionally methyl- or ethyl-substituted pyrrolidinyl, morpholinyl, piperidinyl, piperazinyl or hexahydroazepinyl,

10  $R^4$  represents hydrogen or represents methyl, ethyl, n- or i-propyl, n-, i-, s- or t-butyl, optionally substituted by hydroxyl, formyloxy, phenylcarbonyloxy which is optionally substituted in the phenyl moiety, methoxy, ethoxy, methylthio, ethylthio, methoxycarbonyl, ethoxycarbonyl, methylcarbonyloxy, ethylcarbonyloxy, propylcarbonyloxy, pentylcarbonyloxy or hexylcarbonyloxy, or represents  
15 phenyl, benzyl, 1-phenethyl, 2-phenethyl or indolylmethyl, each of which is optionally substituted in the phenyl moiety or heterocyclyl moiety, or

20  $R^2$  and  $R^4$  together with the atoms to which they are attached represent a pyrrolidine or piperidine ring,

$R^5$  represents hydrogen, methyl, ethyl, n- or i-propyl, n-, i-, s- or t-butyl or

25  $R^4$  and  $R^5$  together with the carbon atom to which they are attached represent a cyclopropane ring, cyclopentane or cyclohexane ring,

30  $R^6$  represents hydrogen, methyl, ethyl, n- or i-propyl, n-, i-, s- or t-butyl, pentyl, hexyl, heptyl, octyl, optionally methyl-, ethyl-, n- or i-propyl-, n-, i-, s- or t-butyl-substituted cyclopentyl or cyclohexyl, or represents phenyl, benzyl, 1-phenethyl, 2-phenethyl, phenylpropyl, phenylbutyl, phenylpentyl or phenylhexyl, pyrrolidinyl, morpholinyl, pyrro-



lidinylbutyl or morpholinylbutyl, each of which is optionally substituted in the phenyl or heterocyclyl moiety, or represents pyrrolidonyl-substituted methyl, ethyl or propyl,

5  $R^7$  represents hydrogen, methyl, ethyl, n- or i-propyl, n-, i-, s- or t-butyl,

$R^8$  represents hydrogen, methyl, ethyl, n- or i-propyl, n-, i-, s- or t-butyl and

10 Z represents hydrogen, methyl, ethyl, n- or i-propyl, n-, i-, s- or t-butyl, pentyl, hexyl, heptyl, octyl, methylcarbonyl, ethylcarbonyl, n- or i-propylcarbonyl, n-, i-, s- or t-butylcarbonyl, pentylcarbonyl, hexylcarbonyl, heptylcarbonyl, octylcarbonyl, optionally methyl-, ethyl-, n- or i-propyl, n-, i-, s- or t-butyl-substituted cyclopentyl, cyclohexyl, cyclopentylcarbonyl or cyclohexylcarbonyl, represents  
15 phenyl, benzyl, 1-phenethyl, 2-phenethyl, phenylpropyl, phenylbutyl, phenylpentyl or phenylhexyl, pyrrolidinyll, morpholinyl, pyrrolidinylbutyl, morpholinylbutyl, phenylcarbonyl, benzylcarbonyl, 1-phenethylcarbonyl, 2-phenethylcarbonyl, phenylcarbonyl-propylcarbonyl, phenylcarbonylbutylcarbonyl, phenylcarbonyl-pentylcarbonyl or phenylcarbonylhexylcarbonyl, pyrrolidinylcarbonyl, morpholinylcarbonyl, pyrrolidinylcarbonylbutylcarbonyl or morpholinylcarbonylbutylcarbonyl, each of which is optionally substituted in the phenyl or heterocyclyl moiety.

25 Particularly preferred substituents for phenyl are given in the list below:

fluorine, chlorine, bromine, cyano, amino, hydroxyl, formyl, carboxyl, carbamoyl, thiocarbamoyl, methyl, ethyl, n- or i-propyl, n-, i-, s- or t-butyl, methoxy, ethoxy, n- or i-propoxy, methylthio, ethylthio, n- or i-propylthio, methylsulphinyl, ethyl-  
30 sulphinyl, methylsulphonyl or ethylsulphonyl, trifluoromethyl, trifluoroethyl, difluoromethoxy, trifluoromethoxy, difluorochloromethoxy, trifluoroethoxy, difluoromethylthio, difluorochloromethylthio, trifluoromethylthio, trifluoromethyl-

5 sulphinyl or trifluoromethylsulphonyl, acetylamino, formylamino, N-formyl-N-methylamino, methylamino, ethylamino, n- or i-propylamino, dimethylamino, diethylamino, acetyl, propionyl, acetyloxy, methoxycarbonyl, ethoxycarbonyl, methylsulphonyloxy, ethylsulphonyloxy, hydroxyiminomethyl, hydroxyiminoethyl, methoxyiminomethyl, ethoxyiminomethyl, methoxyiminoethyl or ethoxyiminoethyl,

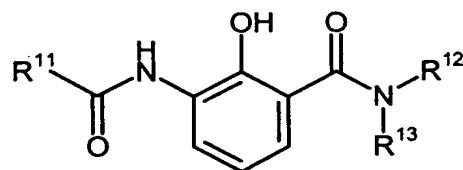
10 in each case doubly attached trimethylene (propane-1,3-diyl), tetramethylene (butane-1,4-diyl), methylenedioxy or ethylenedioxy, each of which is mono- to tetrasubstituted by identical or different substituents from the group consisting of fluorine, chlorine, methyl, trifluoromethyl, ethyl, n- and i-propyl,

cyclopropyl, cyclobutyl, cyclopentyl, cyclohexyl, phenyl or phenoxy.

15 Particularly preferred substituents for heterocyclyl or heterocyclalkyl are given in the list below:

20 halogen, amino, hydroxyl, oxo, methyl, ethyl, n- or i-propyl, n-, i-, s- or t-butyl, methoxy, ethoxy, n- or i-propoxy, methylthio, ethylthio, n- or i-propylthio, methylamino, ethylamino, n- or i-propylamino, dimethylamino or diethylamino.

The present invention furthermore relates to novel substituted acylaminosalicylamides of the general formula (I-a),

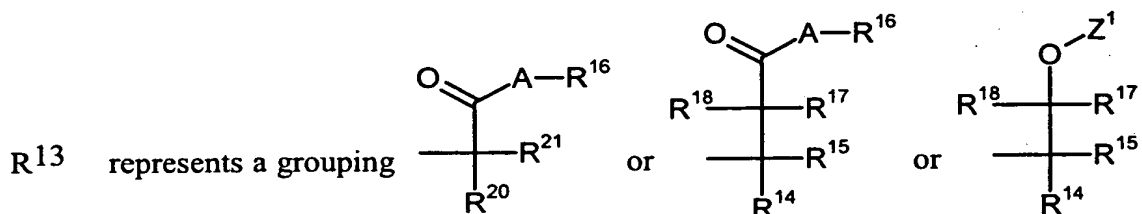


(I-a)

25 in which

R<sup>11</sup> represents hydrogen or alkyl,

R<sup>12</sup> represents hydrogen or alkyl, or



in which

5            A represents oxygen, sulphur or  $\text{---}(\text{N-R}^{19})\text{---}$  in which

R<sup>19</sup> represents hydrogen or alkyl or together with R<sup>16</sup> and the nitrogen atom to which they are attached forms an optionally substituted heterocyclic ring,

10

R<sup>14</sup> represents hydrogen, optionally substituted alkyl or optionally substituted aryl or

R<sup>12</sup> and R<sup>14</sup> together with the atoms to which they are attached form a heterocyclic ring,

15

R<sup>15</sup> represents hydrogen or alkyl or

R<sup>14</sup> and R<sup>15</sup> together with the carbon atom to which they are attached form a carbocyclic ring,

20

R<sup>16</sup> represents hydrogen or in each case optionally substituted alkyl, cycloalkyl, aryl or heterocyclyl,

25

R<sup>17</sup> represents hydrogen or alkyl and

R<sup>18</sup> represents hydrogen or alkyl,

$Z^1$  represents hydrogen or in each case optionally substituted alkyl, alkylcarbonyl, cycloalkyl, cycloalkylcarbonyl, aryl, arylcarbonyl, heterocyclyl or heterocyclcarbonyl,

$R^{20}$  represents hydrogen, optionally substituted alkyl or optionally substituted aryl or hetaryl or

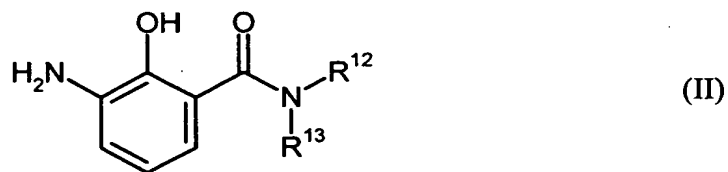
$R^{12}$  and  $R^{20}$  together with the atoms to which they are attached form a heterocyclic ring,

$R^{21}$  represents hydrogen or alkyl or

$R^{20}$  and  $R^{21}$  together with the carbon atom to which they are attached form a carbocyclic ring.

Furthermore, it has been found that the novel substituted acylaminosalicylamides of the general formula (I-a) are obtained when

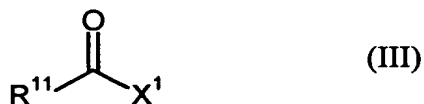
a) aminosalicylamides of the general formula (II),



in which

$R^{12}$  and  $R^{13}$  are each as defined above,

are reacted with an acylating agent of the general formula (III),



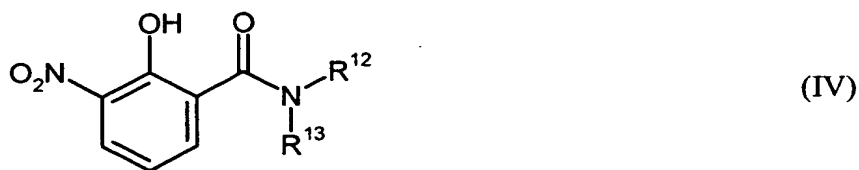
in which

$\text{R}^{11}$  is as defined above and

$\text{X}^1$  represents halogen, hydroxyl, alkoxy or alkylcarbonyloxy,

if appropriate in the presence of a diluent, if appropriate in the presence of an acid acceptor, and if appropriate in the presence of another reaction auxiliary, or when

b) nitrosalicylamides of the general formula (IV)



in which

$\text{R}^{12}$  and  $\text{R}^{13}$  are each as defined above,

are reacted with formic acid, if appropriate in the presence of a catalyst and if appropriate in the presence of a further reaction auxiliary.

Finally, it has been found that the novel acylaminosalicylamides of the general formula (Ia) have strong activity against plant- and material-damaging organisms, in particular very strong fungicidal action. At certain concentrations and applications, the active compounds according to the invention may also be active against plant and animal pests.

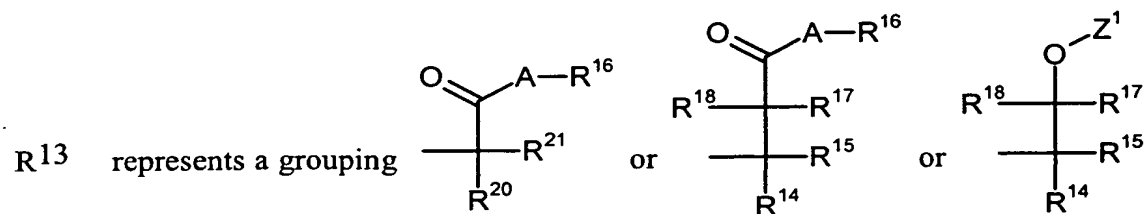
Harmful organisms are to be understood as meaning, in particular, microorganisms.

If appropriate, the compounds according to the invention are present as mixtures of various possible isomeric forms, in particular of stereoisomers, such as, for example, E and Z, threo and erythro, and also optical isomers. What is claimed are both the E and the Z isomers, and also the threo and erythro and the optical isomers, and any mixtures of these isomers.

Preference is given to the novel compounds of the formula (I-a), in which

R<sup>11</sup> represents hydrogen or methyl,

R<sup>12</sup> represents hydrogen or C<sub>1</sub>-C<sub>4</sub>-alkyl and



in which

A represents oxygen, sulphur or -(N-R<sup>19</sup>)- in which

R<sup>19</sup> represents hydrogen or alkyl having 1 to 4 carbon atoms or together with R<sup>16</sup> and the nitrogen atom to which they are attached forms an optionally C<sub>1</sub>-C<sub>4</sub>-alkyl-substituted heterocyclic ring having from 3 to 7 ring members,

R<sup>14</sup> represents hydrogen or alkyl which is optionally substituted by alkoxy, alkylthio, alkoxycarbonyl or alkylcarbonyloxy having in each case 1 to 6 carbon atoms in the alkyl moiety or by arylcarbonyloxy which is optionally substituted in the aryl moiety by hydroxyl, formyloxy, or

represents aryl, heterocyclyl, arylalkyl or heterocyclylalkyl having in each case 1 to 6 carbon atoms in the alkyl moiety and being in each case optionally substituted in the aryl moiety or heterocyclyl moiety, or

5           R<sup>12</sup> and R<sup>14</sup> together with the atoms to which they are attached form a heterocyclic ring having 3 to 6 ring members,

R<sup>15</sup> represents hydrogen or C<sub>1</sub>-C<sub>4</sub>-alkyl or

10           R<sup>14</sup> and R<sup>15</sup> together with the carbon atom to which they are attached form a carbocyclic ring having 3 to 6 ring members,

R<sup>16</sup> represents hydrogen or C<sub>1</sub>-C<sub>12</sub>-alkyl, optionally C<sub>1</sub>-C<sub>4</sub>-alkyl-substituted C<sub>3</sub>-C<sub>7</sub>-cycloalkyl, represents aryl, arylalkyl having 1 to 6 carbon atoms  
15           in the alkyl moiety, heterocyclyl, heterocyclylalkyl having 1 to 6 carbon atoms in the alkyl moiety, each of which is optionally substituted in the aryl or heterocyclyl moiety, or represents pyrrolidonyl-substituted C<sub>1</sub>-C<sub>4</sub>-alkyl,

20           R<sup>17</sup> represents hydrogen or C<sub>1</sub>-C<sub>4</sub>-alkyl and

R<sup>18</sup> represents hydrogen or C<sub>1</sub>-C<sub>4</sub>-alkyl,

Z<sup>1</sup> represents hydrogen or C<sub>1</sub>-C<sub>12</sub>-alkyl or alkylcarbonyl, optionally  
25           C<sub>1</sub>-C<sub>4</sub>-alkyl-substituted C<sub>3</sub>-C<sub>7</sub>-cycloalkyl or cycloalkylcarbonyl, represents aryl, arylcarbonyl, arylalkyl, arylalkylcarbonyl having 1 to 6 carbon atoms in the alkyl moiety, heterocyclyl, heterocyclylcarbonyl, heterocyclylalkyl or heterocyclylalkylcarbonyl having 1 to 6 carbon  
30           atoms in the alkyl moiety, each of which is optionally substituted in the aryl or heterocyclyl moiety,

R<sup>20</sup> represents hydrogen or C<sub>1</sub>-C<sub>4</sub>-alkyl which is optionally substituted by formyloxy, by arylcarbonyloxy which is optionally substituted in the aryl moiety or by alkoxy, alkylthio, alkoxycarbonyl or alkylcarbonyloxy having in each case 1 to 6 carbon atoms in the alkyl moiety or represents aryl, heterocyclyl, arylalkyl having 2 to 6 carbon atoms in the alkyl moiety or heterocyclylalkyl having 1 to 6 carbon atoms in the alkyl moiety, each of which is optionally substituted in the aryl moiety or heterocyclyl moiety, or represents substituted benzyl, or

R<sup>12</sup> and R<sup>20</sup> together with the atoms to which they are attached form a heterocyclic ring having 3 to 6 ring members,

R<sup>21</sup> represents hydrogen or C<sub>1</sub>-C<sub>4</sub>-alkyl or

R<sup>20</sup> and R<sup>21</sup> together with the carbon atom to which they are attached form a carbocyclic ring having 3 to 6 ring members.

Preferred substituents for aryl or arylalkyl are given in the list below:

halogen, cyano, amino, hydroxyl, formyl, carboxyl, carbamoyl, thiocarbamoyl;

in each case straight-chain or branched alkyl, alkoxy, alkylthio, alkylsulphinyl or alkylsulphonyl having in each case 1 to 6 carbon atoms;

in each case straight-chain or branched alkenyl or alkenyloxy having in each case 2 to 6 carbon atoms;

in each case straight-chain or branched halogenoalkyl, halogenoalkoxy, halogenoalkylthio, halogenoalkylsulphinyl or halogenoalkylsulphonyl having in each case 1 to 6 carbon atoms and 1 to 13 identical or different halogen atoms;



in each case straight-chain or branched halogenoalkenyl or halogenoalkenyloxy having in each case 2 to 6 carbon atoms and 1 to 13 identical or different halogen atoms;

5

in each case straight-chain or branched alkylamino, dialkylamino, alkylcarbonyl, alkylcarbonyloxy, alkoxycarbonyl, alkylsulphonyloxy, hydroxyiminoalkyl or alkoxyiminoalkyl having in each case 1 to 6 carbon atoms in the individual alkyl moieties;

10

in each case doubly attached alkylene or dioxyalkylene having in each case 1 to 6 carbon atoms and being in each case optionally mono- or polysubstituted by identical or different substituents selected from the group consisting of halogen, straight-chain or branched alkyl having 1 to 4 carbon atoms and straight-chain or branched halogenoalkyl having 1 to 4 carbon atoms and 1 to 9 identical or different halogen atoms; and

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cycloalkyl having 3 to 6 carbon atoms, aryl and aryloxy.

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Preferred substituents for heterocyclyl or heterocyclylalkyl are given in the list below:

halogen, amino, hydroxyl, oxo,

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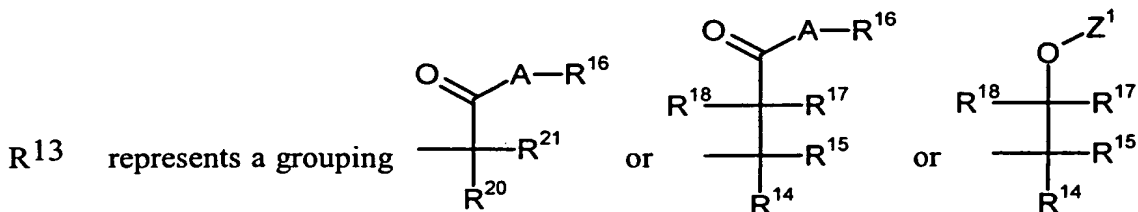
alkyl, alkoxy, alkylthio, alkylamino, dialkylamino having in each case 1 to 6 carbon atoms in the individual alkyl moieties.

The invention relates in particular to the novel compounds of the formula (I-a) in which

30

R<sup>11</sup> represents hydrogen or methyl,

R<sup>12</sup> represents hydrogen, methyl, ethyl, n- or i-propyl, n-, i-, s- or t-butyl and



in which

A represents oxygen, sulphur or  $\text{---}(\text{N} \text{---} \text{R}^{19}) \text{---}$  in which

R<sup>19</sup> represents hydrogen or methyl, ethyl, n- or i-propyl, n-, i-, s- or t-butyl or together with R<sup>16</sup> and the nitrogen atom to which they are attached represents optionally methyl- or ethyl-substituted pyrrolidinyl, morpholinyl, piperidinyl, piperazinyl or hexahydroazepinyl,

R<sup>14</sup> represents hydrogen or represents methyl, ethyl, n- or i-propyl, n-, i-, s- or t-butyl, optionally substituted by hydroxyl, formyloxy, phenylcarbonyloxy which is optionally substituted in the phenyl moiety, methoxy, ethoxy, methylthio, ethylthio, methoxycarbonyl, ethoxycarbonyl, methylcarbonyloxy, ethylcarbonyloxy, propylcarbonyloxy, pentylcarbonyloxy or hexylcarbonyloxy, or represents phenyl, benzyl, 1-phenethyl, 2-phenethyl or indolylmethyl, each of which is optionally substituted in the phenyl moiety or heterocyclyl moiety, or

R<sup>12</sup> and R<sup>14</sup> together with the atoms to which they are attached represent a pyrrolidine or piperidine ring,

R<sup>15</sup> represents hydrogen, methyl, ethyl, n- or i-propyl, n-, i-, s- or t-butyl or

R<sup>14</sup> and R<sup>15</sup> together with the carbon atom to which they are attached represents a cyclopropane ring, cyclopentane or cyclohexane ring,

5 R<sup>16</sup> represents hydrogen, methyl, ethyl, n- or i-propyl, n-, i-, s- or t-butyl, pentyl, hexyl, heptyl, octyl, optionally methyl-, ethyl-, n- or i-propyl-, n-, i-, s- or t-butyl-substituted cyclopentyl or cyclohexyl, or represents phenyl, benzyl 1-phenethyl, 2-phenethyl, phenylpropyl, phenylbutyl, phenylpentyl or phenylhexyl, pyrrolidinyl, morpholinyl, pyrrolidinylbutyl or morpholinylbutyl, each of which is optionally substituted  
10 in the phenyl or heterocyclyl moiety, or represents pyrrolidonyl-substituted methyl, ethyl or propyl,

R<sup>17</sup> represents hydrogen, methyl, ethyl, n- or i-propyl, n-, i-, s- or t-butyl and

15 R<sup>18</sup> represents hydrogen, methyl, ethyl, n- or i-propyl, n-, i-, s- or t-butyl,

Z<sup>1</sup> represents hydrogen, methyl, ethyl, n- or i-propyl, n-, i-, s- or t-butyl, pentyl, hexyl, heptyl, octyl, methylcarbonyl, ethylcarbonyl, n- or i-propylcarbonyl, n-, i-, s- or t-butylcarbonyl, pentylcarbonyl, hexylcarbonyl, heptylcarbonyl, octylcarbonyl, optionally methyl-, ethyl-, n- or i-propyl-, n-, i-, s- or t-butyl-substituted cyclopentyl, cyclohexyl, cyclopentylcarbonyl or cyclohexylcarbonyl, represents phenyl, benzyl, 1-phenethyl, 2-phenethyl, phenylpropyl, phenylbutyl, phenylpentyl or phenylhexyl, pyrrolidinyl, morpholinyl, pyrrolidinylbutyl, morpholinylbutyl, phenylcarbonyl, benzylcarbonyl, 1-phenethylcarbonyl, 2-phenethylcarbonyl, phenylcarbonyl-propylcarbonyl, phenylcarbonylbutylcarbonyl, phenylcarbonylpentylcarbonyl or phenylcarbonylhexylcarbonyl, pyrrolidinylcarbonyl, morpholinylcarbonyl, pyrrolidinylcarbonylbutylcarbonyl or  
20  
25  
30

morpholinylcarbonylbutylcarbonyl, each of which is optionally substituted in the phenyl or heterocyclyl moiety,

5 R<sup>20</sup> represents hydrogen or represents methyl, ethyl, n- or i-propyl, n-, i-, s- or t-butyl, optionally substituted by formyloxy, by phenylcarbonyloxy which is optionally substituted in the phenyl moiety, by methoxy, ethoxy, methylthio, ethylthio, methoxycarbonyl, ethoxycarbonyl, methylcarbonyloxy, ethylcarbonyloxy, propylcarbonyloxy, pentylcarbonyloxy or hexylcarbonyloxy, or represents phenyl, 1-phenethyl, 2-phenethyl or indolylmethyl, each of which is optionally substituted in the phenyl moiety or heterocyclyl moiety, or represents substituted benzyl, or

10 R<sup>12</sup> and R<sup>20</sup> together with the atoms to which they are attached represent a pyrrolidine or piperidine ring,

R<sup>21</sup> represents hydrogen, methyl, ethyl, n- or i-propyl, n-, i-, s- or t-butyl or

20 R<sup>20</sup> and R<sup>21</sup> together with the carbon atom to which they are attached represent a cyclopropane ring, cyclopentane or cyclohexane ring.

Particularly preferred substituents for phenyl are given in the list below:

25 fluorine, chlorine, bromine, cyano, amino, hydroxyl, formyl, carboxyl, carbamoyl, thiocarbamoyl, methyl, ethyl, n- or i-propyl, n-, i-, s- or t-butyl, methoxy, ethoxy, n- or i-propoxy, methylthio, ethylthio, n- or i-propylthio, methylsulphinyl, ethylsulphinyl, methylsulphonyl or ethylsulphonyl, trifluoromethyl, trifluoroethyl, difluoromethoxy, trifluoromethoxy, difluorochloromethoxy, trifluoroethoxy, difluoromethylthio, difluorochloromethylthio, trifluoromethylthio, trifluoromethylsulphinyl or trifluoromethylsulphonyl, acetyl amino, formyl amino, N-formyl-N-methyl amino, methyl amino, ethyl amino, n- or i-propyl amino, dimethyl amino,

30

diethylamino, acetyl, propionyl, acetyloxy, methoxycarbonyl, ethoxycarbonyl, methylsulphonyloxy, ethylsulphonyloxy, hydroxyiminomethyl, hydroxyiminoethyl, methoxyiminomethyl, ethoxyiminomethyl, methoxyiminoethyl or ethoxyiminoethyl,

5 in each case doubly attached trimethylene (propane-1,3-diyl), tetramethylene (butane-1,4-diyl), methylenedioxy or ethylenedioxy, each of which is mono- to tetrasubstituted by identical or different substituents from the group consisting of fluorine, chlorine, methyl, trifluoromethyl, ethyl, n- and i-propyl,

10 cyclopropyl, cyclobutyl, cyclopentyl, cyclohexyl, phenyl or phenoxy.

Particularly preferred substituents for heterocyclyl or heterocyclalkyl are given in the list below:

15 halogen, amino, hydroxyl, oxo, methyl, ethyl, n- or i-propyl, n-, i-, s- or t-butyl, methoxy, ethoxy, n- or i-propoxy, methylthio, ethylthio, n- or i-propylthio, methylamino, ethylamino, n- or i-propylamino, dimethylamino or diethylamino.

20 The abovementioned general or preferred radical definitions apply both to the end products of the formula (I) and, correspondingly, to the starting materials or intermediates required in each case for the preparation.

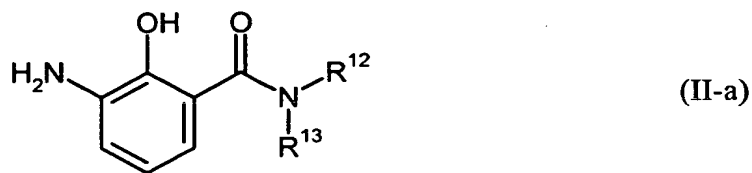
25 The specific radical definitions given in the respective combinations or preferred combinations of radicals are, independently of one another and of the particular combinations of radicals given, also replaced as desired by radical definitions of other radicals.

30 The formula (II) provides a general definition of the aminosalicylamides required as starting materials for carrying out process a) according to the invention. In this formula (II),  $R^{12}$  and  $R^{13}$  each preferably or in particular have those meanings which have already been mentioned in connection with the description of the

compounds of the formula (I) according to the invention as being preferred or as being particularly preferred for R<sup>12</sup> and R<sup>13</sup>.

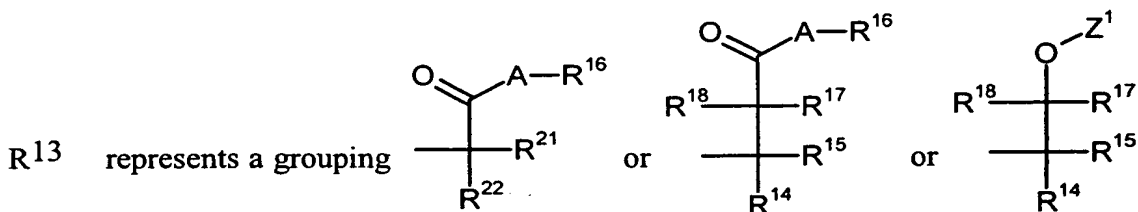
Some of the starting materials of the formula (II) are known (compare, for example, J. Heterocycl. Chem. (1971), 8(6), 989-91).

Novel, and also part of the subject-matter of the present application, are aminosalicylamides of the formula (II-a),



in which

R<sup>12</sup> is as defined above and



A, R<sup>14</sup>, R<sup>15</sup>, R<sup>16</sup>, R<sup>17</sup>, R<sup>18</sup>, Z<sup>1</sup> and R<sup>21</sup> are each as defined above,

R<sup>22</sup> represents C<sub>1</sub>-C<sub>4</sub>-alkyl which is substituted by formyloxy, by arylcarbonyloxy which is optionally substituted in the aryl moiety or by alkoxy, alkylthio, alkoxycarbonyl or alkylcarbonyloxy having in each case 1 to 6 carbon atoms in the alkyl moiety, or represents unsubstituted C<sub>2</sub>-C<sub>4</sub>-alkyl, represents aryl, heterocyclyl, arylalkyl having 2 to 6 carbon atoms in the alkyl moiety or heterocyclylalkyl having 1 to 6

carbon atoms in the alkyl moiety, each of which is optionally substituted in the aryl moiety or heterocyclyl moiety, or represents substituted benzyl, or

5  $R^{22}$  and  $R^{12}$  together with the atoms to which they are attached form a heterocyclic ring,

$R^{22}$  and  $R^{21}$  together with the carbon atom to which they are attached form a carbocyclic ring.

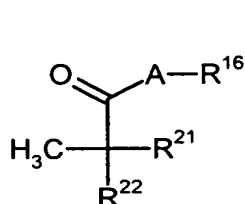
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Preference is given to aminosalicylamides of the formula (II-a) in which

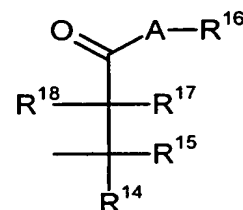
$R^{12}$  is as defined above and

15

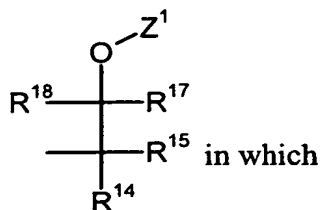
$R^{13}$  represents a grouping



or



or



$A, R^{14}, R^{15}, R^{16}, R^{17}, R^{18}, Z^1$  and  $R^{21}$  are each as defined above,

20

$R^{22}$  represents  $C_1$ - $C_4$ -alkyl which is substituted by formyloxy, by arylcarbonyloxy which is optionally substituted in the aryl moiety or by alkoxy, alkylthio, alkoxycarbonyl or alkylcarbonyloxy having in each case 1 to 6 carbon atoms in the alkyl moiety, or represents unsubstituted  $C_2$ - $C_4$ -alkyl, represents aryl, heterocyclyl, arylalkyl having 2 to 6

carbon atoms in the alkyl moiety or heterocyclalkyl having 1 to 6 carbon atoms in the alkyl moiety, each of which is optionally substituted in the aryl moiety or heterocycl moiety, or represents substituted benzyl, or

5

$R^{22}$  and  $R^{12}$  together with the atoms to which they are attached represent a pyrrolidine or piperidine ring or

$R^{22}$  and  $R^{21}$  together with the carbon atom to which they are attached represent a cyclopentane or cyclohexane ring.

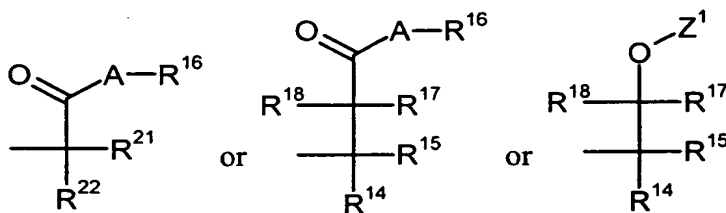
10

Preference is given to aminosalicylamides of the formula (II-a), in which

$R^{12}$  is as defined above and

15

$R^{13}$  represents a grouping



in which

$A$ ,  $R^{14}$ ,  $R^{15}$ ,  $R^{16}$ ,  $R^{17}$ ,  $R^{18}$ ,  $Z^1$  and  $R^{21}$  are each as defined above,

20

$R^{22}$  represents methyl, ethyl, n- or i-propyl, n-, i-, s- or t-butyl, each of which is substituted by formyloxy, by phenylcarbonyloxy which is optionally substituted in the phenyl moiety, by methoxy, ethoxy, methylthio, ethylthio, methoxycarbonyl, ethoxycarbonyl, methylcarbonyloxy, ethylcarbonyloxy, propylcarbonyloxy, pentylcarbonyloxy or hexylcarbonyloxy, or represents unsubstituted ethyl, n- or i-propyl, n-, i-, s- or t-butyl, represents phenyl, 1-phenethyl, 2-phenethyl or indolyl-

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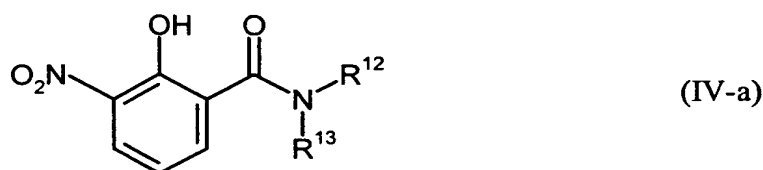


methyl, each of which is optionally substituted in the phenyl moiety or heterocyclyl moiety, or represents substituted benzyl, or

5  $R^{22}$  and  $R^{12}$  together with the atoms to which they are attached represent a pyrrolidine or piperidine ring or

$R^{22}$  and  $R^{21}$  together with the carbon atom to which they are attached represent a cyclopentane or cyclohexane ring.

10 The aminosalicylamides of the formula (II-a) are obtained when (process c) nitrosalicylamides of the general formula (IV-a),



in which

15  $R^{12}$  and  $R^{13}$  are each as defined above

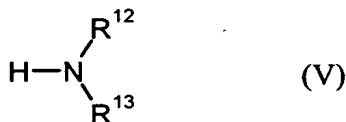
are reacted with hydrogen, if appropriate in the presence of a diluent, preferably an ester, such as methyl acetate or ethyl acetate; an alcohol, such as methanol, ethanol, 20 n- or i-propanol, n-, i-, sec- or tert-butanol, ethanediol, propane-1,2-diol, ethoxyethanol, methoxyethanol, diethylene glycol monomethyl ether, diethylene glycol monoethyl ether; water, a solution of a salt, such as, for example, ammonium chloride solution, an acid, such as, for example, hydrochloric acid or acetic acid, and any mixtures of the diluents mentioned, and if appropriate in the presence of a 25 catalyst, such as, for example, Raney nickel, palladium or platinum, if appropriate on a support, such as activated carbon.

The formula (IV-a) provides a general definition of the nitrosalicylamides required as starting materials for carrying out the process c) according to the invention. In this

formula (IV-a),  $R^{12}$  and  $R^{13}$  each preferably or in particular have those meanings which have already been mentioned in connection with the description of the compounds of the formula (II-a) according to the invention as being preferred or as being particularly preferred for  $R^{12}$  and  $R^{13}$ .

The nitrosalicylamides of the formula (IV-a) are novel and also form part of the subject-matter of the present application.

They are obtained when (process d) 2-hydroxy-3-nitrobenzoic acid or 2-hydroxy-3-nitrobenzoyl chloride are reacted with amine of the formula (V)



in which

$R^{12}$  and  $R^{13}$  are each as defined above,

if appropriate in the presence of a diluent, by way of example and by way of preference an aliphatic, alicyclic or aromatic hydrocarbon, such as, for example, petroleum ether, hexane, heptane, cyclohexane, methylcyclohexane, benzene, toluene, xylene or decalin; a halogenated hydrocarbon, such as, for example, chlorobenzene, dichlorobenzene, dichloromethane, chloroform, carbon tetrachloride, dichloroethane or trichloroethane; an ether, such as, for example, diethyl ether, diisopropyl ether, methyl t-butyl ether, methyl t-amyl ether, dioxane, tetrahydrofuran, 1,2-dimethoxyethane, 1,2-diethoxyethane or anisole; a ketone, such as, for example, acetone, butanone, methyl isobutyl ketone or cyclohexanone; a nitrile, such as, for example, acetonitrile, propionitrile, n- or i-butyronitrile or benzonitrile; an amide, such as, for example, N,N-dimethylformamide, N,N-dimethylacetamide, N-methylformanilide, N-methylpyrrolidone or hexamethylphosphoric triamide; an ester, such as, for example, methyl acetate or ethyl acetate; a sulfoxide, such as, for example, dimethylsulphoxide; or a sulphone, such as, for example, sulfolane, if

appropriate in the presence of a condensing agent, for example an acyl halide former, such as phosgene, phosphorus tribromide, phosphorus trichloride, phosphorus pentachloride, phosphorus oxychloride or thionylchloride; an anhydride former, such as, for example, ethyl chloroformate, methyl chloroformate, isopropyl chloroformate, isobutyl chloroformate or methanesulphonyl chloride; a carbodiimide, such as, for example, N,N'-dicyclohexylcarbodiimide (DCC), or another customary condensing agent, such as, for example, phosphorus pentoxide, polyphosphoric acid, N,N'-carbonyldiimidazole, 2-ethoxy-N-ethoxycarbonyl-1,2-dihydroquinoline (EEDQ) or triphenylphosphine/carbon tetrachloride, and if appropriate in the presence of an acid acceptor, by way of example and by way of preference an alkaline earth metal or alkali metal hydride, hydroxide, amide, alkoxide, acetate, carbonate or bicarbonate, such as, for example, sodium hydride, sodium amide, sodium methoxide, sodium ethoxide, potassium tert-butoxide, sodium hydroxide, potassium hydroxide, ammonium hydroxide, sodium acetate, potassium acetate, calcium acetate, ammonium acetate, sodium carbonate, potassium carbonate, potassium bicarbonate, sodium bicarbonate or ammonium carbonate, or a tertiary amine, such as, for example, trimethylamine, triethylamine, tributylamine, N,N-dimethylaniline, N,N-dimethylbenzylamine, pyridine, N-methylpiperidine, N-methylmorpholine, N,N-dimethylaminopyridine, diazabicyclooctane (DABCO), diazabicyclononene (DBN) or diazabicycloundecene (DBU).

2-Hydroxy-3-nitrobenzoic acid and 2-hydroxy-3-nitrobenzoyl chloride, required as starting materials for carrying out the process d) according to the invention, are known (compare, for example, J. Het. Chem., 1971, 8(6), 889-891, J.Chem.Soc., 1953, 2049, 2050 or US 03527865).

The formula (V) provides a general definition of the amines furthermore required as starting materials for carrying out the process d) according to the invention. In this formula (V), R<sup>12</sup> and R<sup>13</sup> by way of example and by way of preference or in particular have both meanings which have already been mentioned in connection with the description of the compounds of the formula (I) according to the invention as being preferred or as being particularly preferred for R<sup>12</sup> and R<sup>13</sup>.

The amines of the formula (VII) are known reagents of organic chemistry.

5 Some of the compounds of the formula (I) are known, and they can be prepared by processes, some of which are known (cf. Biochim. Biophys. Acta 1993, 262-268).

10 The formula (III) provides a general definition of the acylating agents furthermore required as starting materials for carrying out the process a) according to the invention. In this formula (III),  $R^{11}$  preferably or in particular has that meaning which has already been mentioned in connection with the description of the compounds of the formula (I) according to the invention as being preferred or as being particularly preferred for  $R^{11}$ .  $X^1$  represents halogen, hydroxyl, alkoxy or alkylcarbonyloxy, preferably chlorine, hydroxyl, methoxy, ethoxy or acetoxy.

15 The acylating agents of the general formula (III) are known reagents of organic chemistry.

20 The formula (IV) provides a general definition of the nitrosalicylamides required as starting materials for carrying out the process b) according to the invention. In the formula (IV),  $R^{12}$  and  $R^{13}$  each preferably or in particular have those meanings which have already been mentioned in connection with the description of the compounds of the formula (I) according to the invention as being preferred or as being particularly preferred for  $R^{12}$  and  $R^{13}$ .

25 Some of the starting materials of the formula (IV) are known (compare, for example, J. Heterocycl. Chem. (1971), 8(6), 989-991).

30 The nitrosalicylamides of the formula (IV-a), which have already been described further above in connection with the description of the process c) according to the invention are novel.

Suitable diluents for carrying out the process a) according to the invention are all inert organic solvents. These include, by way of example and by way of preference, aliphatic, alicyclic or aromatic hydrocarbons such as, for example, petroleum ether, hexane, heptane, cyclohexane, methylcyclohexane, benzene, toluene, xylene or decalin; halogenated hydrocarbons, such as, for example, chlorobenzene, dichlorobenzene, dichloromethane, chloroform, carbon tetrachloride, dichloroethane or trichloroethane; ethers, such as, for example, diethyl ether, diisopropyl ether, methyl t-butyl ether, methyl t-amyl ether, dioxane, tetrahydrofuran, 1,2-dimethoxyethane, 1,2-diethoxyethane or anisole; ketones, such as, for example, acetone, butanone, methyl isobutyl ketone or cyclohexanone; nitriles, such as, for example, acetonitrile, propionitrile, n- or i-butyronitrile or benzonitrile; amides, such as, for example N,N-dimethylformamide, N,N-dimethylacetamide, N-methylformanilide, N-methylpyrrolidone or hexamethylphosphoric triamide; esters, such as, for example, methyl acetate or ethyl acetate; sulphoxides, such as, for example, dimethyl sulphoxide, or sulphones, such as sulpholane.

If appropriate, the process a) according to the invention is carried out in the presence of a suitable acid acceptor. Suitable acid acceptors are all customary inorganic or organic bases. These preferably include alkaline earth metal or alkali metal hydroxides, acetates, carbonates or bicarbonates, such as, for example, sodium hydroxide, potassium hydroxide, sodium acetate, potassium acetate, calcium acetate, sodium carbonate, potassium carbonate, potassium bicarbonate or sodium bicarbonate, and also tertiary amines, such as trimethylamine, triethylamine, tributylamine, N,N-dimethylaniline, N,N-dimethyl-benzylamine, pyridine, N-methylpiperidine, N-methylmorpholine, N,N-dimethylaminopyridine, diazabicyclooctane (DABCO), diazabicyclononene (DBN) or diazabicycloundecene (DBU).

If appropriate, the process b) according to the invention is carried out in the presence of a catalyst. Suitable catalysts are all catalysts which are customarily used for hydrogenations. Examples which may be mentioned are: Raney nickel, palladium or platinum, if appropriate on a support, such as, for example, activated carbon.

When carrying out the process a) and b) according to the invention, the reaction temperatures can be varied within a relatively wide range. In general, the processes are carried out at temperatures from 0°C to 180°C, preferably at temperatures from 0°C to 130°C.

5

For carrying out the process a) according to the invention for preparing compounds of the formula (I), in general from 1 to 2000 mol, preferably from 1 to 800 mol, of acylating agent of the formula (III) are employed per mole of the aminosalicylamide of the formula (II).

10

For carrying out the process b) according to the invention for preparing the compounds of the formula (I), generally from 100 to 2000 mol, preferably from 200 to 1000 mol, of formic acid are employed per mole of the nitrosalicylamide of the formula (IV-a).

15

The processes according to the invention are generally carried out under atmospheric pressure. However, it is also possible to operate under elevated or reduced pressure - generally between 0.1 bar and 10 bar.

20

The compounds according to the invention have potent microbicidal activity and can be employed for controlling undesirable microorganisms, such as fungi and bacteria, in crop protection and in the protection of materials.

25

Fungicides are employed in crop protection for controlling Plasmodiophoromycetes, Oomycetes, Chytridiomycetes, Zygomycetes, Ascomycetes, Basidiomycetes and Deuteromycetes.

30

Bactericides are employed in crop protection for controlling Pseudomonadaceae, Rhizobiaceae, Enterobacteriaceae, Corynebacteriaceae and Streptomycetaceae.

Some pathogens causing fungal and bacterial diseases which come under the generic names listed above are mentioned as examples, but not by way of limitation:

Xanthomonas species, such as, for example, Xanthomonas campestris pv. oryzae;

*Pseudomonas* species, such as, for example, *Pseudomonas syringae* pv. *lachrymans*;

*Erwinia* species, such as, for example, *Erwinia amylovora*;

*Pythium* species, such as, for example, *Pythium ultimum*;

*Phytophthora* species, such as, for example, *Phytophthora infestans*;

5 *Pseudoperonospora* species, such as, for example, *Pseudoperonospora humuli* or  
*Pseudoperonospora cubensis*;

*Plasmopara* species, such as, for example, *Plasmopara viticola*;

*Bremia* species, such as, for example, *Bremia lactucae*;

*Peronospora* species, such as, for example, *Peronospora pisi* or *P. brassicae*;

10 *Erysiphe* species, such as, for example, *Erysiphe graminis*;

*Sphaerotheca* species, such as, for example, *Sphaerotheca fuliginea*;

*Podosphaera* species, such as, for example, *Podosphaera leucotricha*;

*Venturia* species, such as, for example, *Venturia inaequalis*;

*Pyrenophora* species, such as, for example, *Pyrenophora teres* or *P. graminea*  
15 (conidia form: *Drechslera*, syn: *Helminthosporium*);

*Cochliobolus* species, such as, for example, *Cochliobolus sativus*  
(conidia form: *Drechslera*, syn: *Helminthosporium*);

*Uromyces* species, such as, for example, *Uromyces appendiculatus*;

*Puccinia* species, such as, for example, *Puccinia recondita*;

20 *Sclerotinia* species, such as, for example, *Sclerotinia sclerotiorum*;

*Tilletia* species, such as, for example, *Tilletia caries*;

*Ustilago* species, such as, for example, *Ustilago nuda* or *Ustilago avenae*;

*Pellicularia* species, such as, for example, *Pellicularia sasakii*;

*Pyricularia* species, such as, for example, *Pyricularia oryzae*;

25 *Fusarium* species, such as, for example, *Fusarium culmorum*;

*Botrytis* species, such as, for example, *Botrytis cinerea*;

*Septoria* species, such as, for example, *Septoria nodorum*;

*Leptosphaeria* species, such as, for example, *Leptosphaeria nodorum*;

*Cercospora* species, such as, for example, *Cercospora canescens*;

30 *Alternaria* species, such as, for example, *Alternaria brassicae*;

*Pseudocercospora* species, such as, for example, *Pseudocercospora*  
*herpotrichoides*.

35 The fact that the active compounds are well tolerated by plants at the concentrations  
required for controlling plant diseases permits the treatment of aerial parts of plants,  
of propagation stock and seeds, and of the soil.

The active compounds according to the invention can be employed particularly successfully for controlling diseases in fruit and vegetable growing and viticulture, such as, for example, against Botrytis, Phytophthora and Plasmopara species or rice diseases, such as, for example, Pyricularia species.

5

According to the invention, it is possible to treat all plants and parts of plants. By plants are to be understood here all plants and plant populations such as desired and undesired wild plants or crop plants (including naturally occurring crop plants). Crop plants can be plants which can be obtained by conventional breeding and optimization methods or by biotechnological and genetic engineering methods or combinations of these methods, including the transgenic plants and including plant cultivars which can or cannot be protected by plant breeder certificates. Parts of plants are to be understood as meaning all above-ground and below-ground parts and organs of plants, such as shoot, leaf, flower and root, examples which may be mentioned being leaves, needles, stems, trunks, flowers, fruit-bodies, fruits and seeds and also roots, tubers and rhizomes. Parts of plants also include harvested plants and vegetative and generative propagation material, for example seedlings, tubers, rhizomes, cuttings and seeds.

10

15

20

The treatment of the plants and parts of plants according to the invention with the active compounds is carried out directly or by action on their environment, habitat or storage area according to customary treatment methods, for example by dipping, spraying, evaporating, atomizing, broadcasting, brushing-on and, in the case of propagation material, in particular in the case of seeds, furthermore by one- or multi-layer coating.

25

Cereal diseases are likewise controlled successfully.

The active compounds according to the invention are also suitable for increasing the harvest yield. Moreover, they show reduced toxicity and are well tolerated by plants.

30

Depending on their particular physical and/or chemical properties, the active compounds can be converted into the customary formulations, such as solutions, emulsions, suspensions, powders, foams, pastes, granules, aerosols and micro-encapsulations in polymeric substances and in coating compositions for seeds, and ULV cool and warm fogging formulations.

35



These formulations are produced in a known manner, for example by mixing the active compounds with extenders, that is, liquid solvents, liquefied gases under pressure, and/or solid carriers, optionally with the use of surfactants, that is emulsifiers and/or dispersants, and/or foam-formers. If the extender used is water, it is also possible to employ, for example, organic solvents as auxiliary solvents. Essentially, the following are suitable for use as liquid solvents: aromatics such as xylene, toluene or alkyl naphthalenes, chlorinated aromatics or chlorinated aliphatic hydrocarbons such as chlorobenzenes, chloroethylenes or methylene chloride, aliphatic hydrocarbons such as cyclohexane or paraffins, for example petroleum fractions, alcohols such as butanol or glycol and their ethers and esters, ketones such as acetone, methyl ethyl ketone, methyl isobutyl ketone or cyclohexanone, strongly polar solvents such as dimethylformamide or dimethyl sulphoxide, or else water. Liquefied gaseous extenders or carriers are to be understood as meaning liquids which are gaseous at standard temperature and under atmospheric pressure, for example aerosol propellants such as halogenated hydrocarbons, or else butane, propane, nitrogen and carbon dioxide. Suitable solid carriers are: for example ground natural minerals such as kaolins, clays, talc, chalk, quartz, attapulgite, montmorillonite or diatomaceous earth, and ground synthetic minerals such as highly disperse silica, alumina and silicates. Suitable solid carriers for granules are: for example crushed and fractionated natural rocks such as calcite, marble, pumice, sepiolite and dolomite, or else synthetic granules of inorganic and organic meals, and granules of organic material such as sawdust, coconut shells, maize cobs and tobacco stalks. Suitable emulsifiers and/or foam formers are: for example nonionic and anionic emulsifiers, such as polyoxyethylene fatty acid esters, polyoxyethylene fatty alcohol ethers, for example alkylaryl polyglycol ethers, alkylsulphonates, alkyl sulphates, arylsulphonates, or else protein hydrolysates. Suitable dispersants are: for example lignin-sulphite waste liquors and methylcellulose.

Tackifiers such as carboxymethylcellulose and natural and synthetic polymers in the form of powders, granules or latices, such as gum arabic, polyvinyl alcohol and polyvinyl acetate, or else natural phospholipids such as cephalins and lecithins and synthetic phospholipids can be used in the formulations. Other suitable additives are mineral and vegetable oils.

It is possible to use colourants such as inorganic pigments, for example iron oxide, titanium oxide and Prussian Blue, and organic dyestuffs such as alizarin dyestuffs,

azo dyestuffs and metal phthalocyanine dyestuffs, and trace nutrients such as salts of iron, manganese, boron, copper, cobalt, molybdenum and zinc.

5 The formulations generally comprise between 0.1 and 95 per cent by weight of active compound, preferably between 0.5 and 90%.

10 The active compounds according to the invention can be used as such or in their formulations also mixed with known fungicides, bactericides, acaricides, nematocides or insecticides in order thus, for example, to widen the spectrum of action or to prevent development of resistance. In many cases, synergistic effects are achieved, i.e. the activity of the mixture exceeds the activity of the individual components.

Examples of co-components in mixtures are the following compounds:

15 **Fungicides:**

aldimorph, ampropylfos, ampropylfos potassium, andoprim, anilazine, azaconazole, azoxystrobin,  
benalaxyl, benodanil, benomyl, benzamacril, benzamacryl-isobutyl, bialaphos, binapacryl, biphenyl, bitertanol, blasticidin-S, bromuconazole, bupirimate,  
20 buthiobate,  
calcium polysulphide, capsimycin, captafol, captan, carbendazim, carboxin, carvon, quinomethionate, chlobenthiazole, chlorfenazole, chloroneb, chloropicrin, chlorothalonil, chlozolate, clozylacon, cufraneb, cymoxanil, cyproconazole, cyprodinil, cyprofuram,  
25 debacarb, dichlorophen, diclobutrazole, diclofluanid, diclomezine, dicloran, diethofencarb, difenoconazole, dimethirimol, dimethomorph, diniconazole, diniconazole-M, dinocap, diphenylamine, dipyrithione, ditalimfos, dithianon, dodemorph, dodine, drazoxolon, edifenphos, epoxiconazole, etaconazole, ethirimol, etridiazole,  
30 famoxadon, fenapanil, fenarimol, fenbuconazole, fenfuram, fenitropan, fenciclonil, fenpropidin, fenpropimorph, fentin acetate, fentin hydroxide, ferbam, ferimzone, fluazinam, flumetover, fluoromide, fluquinconazole, flurprimidol, flusilazole, flusulfamide, flutolanil, flutriafol, folpet, fosetyl-aluminium, fosetyl-sodium, fthalide, fuberidazole, furalaxyl, furametpyr, furcarbonil, furconazole, furconazole-  
35 cis, furmecyclox,  
guazatine,  
hexachlorobenzene, hexaconazole, hymexazole,

imazalil, imibenconazole, iminoctadine, iminoctadine albesilate, iminoctadine triacetate, iodocarb, ipconazole, iprobenfos (IBP), iprodione, irumamycin, isoprothiolane, isovaledione,

kasugamycin, kresoxim-methyl, copper preparations, such as: copper hydroxide, copper naphthenate, copper oxychloride, copper sulphate, copper oxide, oxine-copper and Bordeaux mixture,

mancopper, mancozeb, maneb, meferimzone, mepanipirim, mepronil, metalaxyl, metconazole, methasulfocarb, methfuroxam, metiram, metomeclam, metsulfovax, mildiomyacin, myclobutanil, myclozolin,

nickel dimethyldithiocarbamate, nitrothal-isopropyl, nuarimol,

ofurace, oxadixyl, oxamocarb, oxolinic acid, oxycarboxim, oxyfenthiin,

paclobutrazole, pefurazoate, penconazole, pencycuron, phosdiphen, pimaricin, piperalin, polyoxin, polyoxorim, probenazole, prochloraz, procymidone, propamocarb, propanosine-sodium, propiconazole, propineb, pyrazophos, pyrifenox,

pyrimethanil, pyroquilon, pyroxyfur,

quinconazole, quinoxifen, quintozone (PCNB),

sulphur and sulphur preparations,

tebuconazole, tecloftalam, tecnazene, tetcyclacis, tetraconazole, thiabendazole, thicyofen, thifluzamide, thiophanate-methyl, thiram, tioxymid, tolclofos-methyl,

tolyfluanid, triadimefon, triadimenol, triazbutil, triazoxide, trichlamide, tricyclazole, tridemorph, triflumizole, triforine, triticonazole,

uniconazole,

validamycin A, vinclozolin, viniconazole,

zarilamide, zineb, ziram and also

Dagger G,

OK-8705,

OK-8801,

$\alpha$ -(1,1-dimethylethyl)- $\beta$ -(2-phenoxyethyl)-1H-1,2,4-triazole-1-ethanol,

$\alpha$ -(2,4-dichlorophenyl)- $\beta$ -fluoro- $\beta$ -propyl-1H-1,2,4-triazole-1-ethanol,

$\alpha$ -(2,4-dichlorophenyl)- $\beta$ -methoxy- $\alpha$ -methyl-1H-1,2,4-triazole-1-ethanol,

$\alpha$ -(5-methyl-1,3-dioxan-5-yl)- $\beta$ -[[4-(trifluoromethyl)-phenyl]-methylene]-1H-1,2,4-triazole-1-ethanol,

(5RS,6RS)-6-hydroxy-2,2,7,7-tetramethyl-5-(1H-1,2,4-triazol-1-yl)-3-octanone,

(E)- $\alpha$ -(methoxyimino)-N-methyl-2-phenoxy-phenylacetamide,

1-isopropyl {2-methyl-1-[[[1-(4-methylphenyl)-ethyl]-amino]-carbonyl]-propyl}-carbamate,

1-(2,4-dichlorophenyl)-2-(1H-1,2,4-triazol-1-yl)-ethanone O-(phenylmethyl)-oxime,

- 1-(2-methyl-1-naphthalenyl)-1H-pyrrole-2,5-dione,  
 1-(3,5-dichlorophenyl)-3-(2-propenyl)-2,5-pyrrolidinedione,  
 1-[(diiodomethyl)-sulphonyl]-4-methyl-benzene,  
 1-[[2-(2,4-dichlorophenyl)-1,3-dioxolan-2-yl]-methyl]-1H-imidazole,  
 5 1-[[2-(4-chlorophenyl)-3-phenyloxiranyl]-methyl]-1H-1,2,4-triazole,  
 1-[1-[2-[(2,4-dichlorophenyl)-methoxy]-phenyl]-ethenyl]-1H-imidazole,  
 1-methyl-5-nonyl-2-(phenylmethyl)-3-pyrrolidinole,  
 2',6'-dibromo-2-methyl-4'-trifluoromethoxy-4'-trifluoro-methyl-1,3-thiazole-5-  
 carboxanilide,  
 10 2,2-dichloro-N-[1-(4-chlorophenyl)-ethyl]-1-ethyl-3-methyl-  
 cyclopropanecarboxamide,  
 2,6-dichloro-5-(methylthio)-4-pyrimidinyl-thiocyanate,  
 2,6-dichloro-N-(4-trifluoromethylbenzyl)-benzamide,  
 2,6-dichloro-N-[[4-(trifluoromethyl)-phenyl]-methyl]-benzamide,  
 15 2-(2,3,3-triiodo-2-propenyl)-2H-tetrazole,  
 2-[(1-methylethyl)sulphonyl]-5-(trichloromethyl)-1,3,4-thiadiazole,  
 2-[[6-deoxy-4-O-(4-O-methyl-β-D-glycopyranosyl)-α-D-glucopyranosyl]-amino]-4-  
 methoxy-1H-pyrrolo[2,3-d]pyrimidine-5-carbonitrile,  
 2-aminobutane,  
 20 2-bromo-2-(bromomethyl)-pentanedinitrile,  
 2-chloro-N-(2,3-dihydro-1,1,3-trimethyl-1H-inden-4-yl)-3-pyridinecarboxamide,  
 2-chloro-N-(2,6-dimethylphenyl)-N-(isothiocyanatomethyl)-acetamide,  
 2-phenylphenol (OPP),  
 3,4-dichloro-1-[4-(difluoromethoxy)-phenyl]-1H-pyrrole-2,5-dione,  
 25 3,5-dichloro-N-[cyano-[(1-methyl-2-propynyl)-oxy]-methyl]-benzamide,  
 3-(1,1-dimethylpropyl-1-oxo-1H-indene-2-carbonitrile),  
 3-[2-(4-chlorophenyl)-5-ethoxy-3-isoxazolidinyl]-pyridine,  
 4-chloro-2-cyano-N,N-dimethyl-5-(4-methylphenyl)-1H-imidazole-1-sulphonamide,  
 4-methyl-tetrazolo[1,5-a]quinazolin-5(4H)-one,  
 30 8-(1,1-dimethylethyl)-N-ethyl-N-propyl-1,4-dioxaspiro[4.5]decane-2-methanamine,  
 8-hydroxyquinoline sulphate,  
 9H-xanthene-2-[(phenylamino)-carbonyl]-9-carboxylic hydrazide,  
 bis-(1-methylethyl)-3-methyl-4-[(3-methylbenzoyl)-oxy]-2,5-  
 thiophenedicarboxylate,  
 35 cis-1-(4-chlorophenyl)-2-(1H-1,2,4-triazol-1-yl)-cycloheptanol,  
 cis-4-[3-[4-(1,1-dimethylpropyl)-phenyl-2-methylpropyl]-2,6-dimethyl-  
 morpholinehydrochloride,

ethyl [(4-chlorophenyl)-azo]-cyanoacetate,  
 potassium hydrogen carbonate,  
 methanetetrahiol sodium salt,  
 methyl 1-(2,3-dihydro-2,2-dimethyl-1H-inden-1-yl)-1H-imidazole-5-carboxylate,  
 5 methyl N-(2,6-dimethylphenyl)-N-(5-isoxazolylcarbonyl)-DL-alaninate,  
 methyl N-(chloroacetyl)-N-(2,6-dimethylphenyl)-DL-alaninate,  
 N-(2,3-dichloro-4-hydroxyphenyl)-1-methyl-cyclohexanecarboxamide,  
 N-(2,6-dimethylphenyl)-2-methoxy-N-(tetrahydro-2-oxo-3-furanyl)-acetamide,  
 N-(2,6-dimethylphenyl)-2-methoxy-N-(tetrahydro-2-oxo-3-thienyl)-acetamide,  
 10 N-(2-chloro-4-nitrophenyl)-4-methyl-3-nitro-benzenesulphonamide,  
 N-(4-cyclohexylphenyl)-1,4,5,6-tetrahydro-2-pyrimidineamine,  
 N-(4-hexylphenyl)-1,4,5,6-tetrahydro-2-pyrimidineamine,  
 N-(5-chloro-2-methylphenyl)-2-methoxy-N-(2-oxo-3-oxazolidinyl)-acetamide,  
 N-(6-methoxy-3-pyridinyl)-cyclopropanecarboxamide,  
 15 N-[2,2,2-trichloro-1-[(chloroacetyl)-amino]-ethyl]-benzamide,  
 N-[3-chloro-4,5-bis(2-propinyloxy)-phenyl]-N'-methoxy-methanimidamide,  
 N-formyl-N-hydroxy-DL-alanine-sodium salt,  
 O,O-diethyl [2-(dipropylamino)-2-oxoethyl]-ethylphosphoramidothioate,  
 O-methyl S-phenyl phenylpropylphosphoramidothioate,  
 20 S-methyl 1,2,3-benzothiadiazole-7-carbothioate,  
 spiro[2H]-1-benzopyrane-2,1'(3'H)-isobenzofuran]-3'-one,

#### **Bactericides:**

bronopol, dichlorophen, nitrapyrin, nickel dimethyldithiocarbamate, kasugamycin,  
 25 othilinone, furancarboxylic acid, oxytetracyclin, probenazole, streptomycin,  
 tecloftalam, copper sulphate and other copper preparations.

#### **Insecticides / acaricides / nematocides:**

abamectin, acephate, acetamiprid, acrinathrin, alanycarb, aldicarb, aldoxycarb, alpha-  
 30 cypermethrin, alphasmethrin, amitraz, avermectin, AZ 60541, azadirachtin,  
 azamethiphos, azinphos A, azinphos M, azocyclotin,  
 Bacillus popilliae, Bacillus sphaericus, Bacillus subtilis, Bacillus thuringiensis,  
 baculoviruses, Beauveria bassiana, Beauveria tenella, bendiocarb, benfuracarb,  
 bensultap, benzoximate, betacyfluthrin, bifenazate, bifenthrin, bioethanomethrin,  
 35 biopermethrin, BPMC, bromophos A, bufencarb, buprofezin, butathiofos,  
 butocarboxim, butylpyridaben,

cadusafos, carbaryl, carbofuran, carbophenothion, carbosulfan, cartap, chloethocarb,  
 chlorethoxyfos, chlorfenapyr, chlorfenvinphos, chlorfluazuron, chlormephos,  
 chlorpyrifos, chlorpyrifos M, chlovaporthrin, cis-resmethrin, cispermethrin,  
 clocythrín, cloethocarb, clofentezine, cyanophos, cycloprene, cycloprothrin,  
 5 cyfluthrin, cyhalothrin, cyhexatin, cypermethrin, cyromazine,  
 deltamethrin, demeton M, demeton S, demeton-S-methyl, diafenthiuron, diazinon,  
 dichlorvos, diflubenzuron, dimethoat, dimethylvinphos, diofenolan, disulfoton,  
 docusat-sodium, dofenapyn,  
 eflusilanate, emamectin, empenthrin, endosulfan, Entomopftthora spp., esfenvalerate,  
 10 ethiofencarb, ethion, ethoprophos, etofenprox, etoxazole, etrimfos,  
 fenamiphos, fenazaquin, fenbutatin oxide, fenitrothion, fenothiocarb, fenoxacrim,  
 fenoxycarb, fenpropathrin, fenpyrad, fenpyrithrin, fenpyroximate, fenvalerate,  
 fipronil, fluazinam, fluazuron, flubrocycythrinate, flucycloxuron, flucythrinate,  
 flufenoxuron, flutenzine, fluvalinate, fonophos, fosmethilan, fosthiazate, fubfenprox,  
 15 furathiocarb,  
 granulosis viruses,  
 halofenozide, HCH, heptenophos, hexaflumuron, hexythiazox, hydroprene,  
 imidacloprid, isazofos, isofenphos, isoxathion, ivermectin,  
 lambda-cyhalothrin, lufenuron,  
 20 malathion, mecarbam, metaldehyde, methamidophos, Metharhizium anisopliae,  
 Metharhizium flavoviride, methidathion, methiocarb, methomyl, methoxyfenozide,  
 metolcarb, metoxadiazone, mevinphos, milbemectin, monocrotophos,  
 naled, nitenpyram, nithiazine, novaluron, nuclear polyhedrosis viruses,  
 omethoat, oxamyl, oxydemethon M,  
 25 Paecilomyces fumosoroseus, parathion A, parathion M, permethrin, phenthoat,  
 phorat, phosalone, phosmet, phosphamidon, phoxim, pirimicarb, pirimiphos A,  
 pirimiphos M, profenofos, promecarb, propoxur, prothiofos, prothoat, pymetrozine,  
 pyraclofos, pyresmethrin, pyrethrum, pyridaben, pyridathion, pyrimidifen,  
 pyriproxyfen,  
 30 quinalphos,  
 ribavirin,  
 salithion, sebufos, silafluofen, spinosad, sulfotep, sulprofos,  
 tau-fluvalinate, tebufenozide, tebufenpyrad, tebupirimiphos, teflubenzuron,  
 tefluthrin, temephos, temevinphos, terbufos, tetrachlorvinphos, theta-cypermethrin,  
 35 thiamethoxam, thiapronil, thiatrphos, thiocyclam hydrogen oxalate, thiodicarb,  
 thiofanox, thuringiensin, tralocycythrín, tralomethrin, triarathene, triazamate,  
 triazophos, triazuron, trichlophenidine, trichlorfon, triflumuron, trimethacarb,

vamidothion, vaniliprole, *Verticillium lecanii*,  
 YI 5302,

zeta-cypermethrin, zolaprofos,

(1R-cis)-[5-(phenylmethyl)-3-furanyl]-methyl-3-[(dihydro-2-oxo-3(2H)-  
 furanylidene)-methyl]-2,2-dimethylcyclopropanecarboxylate,

(3-phenoxyphenyl)-methyl-2,2,3,3-tetramethylcyclopropanecarboxylate,  
 1-[(2-chloro-5-thiazolyl)methyl]tetrahydro-3,5-dimethyl-N-nitro-1,3,5-triazine-  
 2(1H)-imine,

2-(2-chloro-6-fluorophenyl)-4-[4-(1,1-dimethylethyl)phenyl]-4,5-dihydro-oxazole,  
 2-(acetyloxy)-3-dodecyl-1,4-naphthalenedione,

2-chloro-N-[[[4-(1-phenylethoxy)-phenyl]-amino]-carbonyl]-benzamide,  
 2-chloro-N-[[[4-(2,2-dichloro-1,1-difluoroethoxy)-phenyl]-amino]-carbonyl]-  
 benzamide,

3-methylphenyl propylcarbamate

4-[4-(4-ethoxyphenyl)-4-methylpentyl]-1-fluoro-2-phenoxy-benzene,

4-chloro-2-(1,1-dimethylethyl)-5-[[2-(2,6-dimethyl-4-phenoxyphenoxy)ethyl]thio]-  
 3(2H)-pyridazinone,

4-chloro-2-(2-chloro-2-methylpropyl)-5-[(6-iodo-3-pyridinyl)methoxy]-3(2H)-  
 pyridazinone,

4-chloro-5-[(6-chloro-3-pyridinyl)methoxy]-2-(3,4-dichlorophenyl)-3(2H)-  
 pyridazinone,

*Bacillus thuringiensis* strain EG-2348,

[2-benzoyl-1-(1,1-dimethylethyl)-hydrazinobenzoic acid,

2,2-dimethyl-3-(2,4-dichlorophenyl)-2-oxo-1-oxaspiro[4.5]dec-3-en-4-yl butanoate,

[3-[(6-chloro-3-pyridinyl)methyl]-2-thiazolidinylidene]-cyanamide,

dihydro-2-(nitromethylene)-2H-1,3-thiazine-3(4H)-carboxaldehyde,

ethyl [2-[[1,6-dihydro-6-oxo-1-(phenylmethyl)-4-pyridazinyl]oxy]ethyl]-carbamate,

N-(3,4,4-trifluoro-1-oxo-3-butenyl)-glycine,

N-(4-chlorophenyl)-3-[4-(difluoromethoxy)phenyl]-4,5-dihydro-4-phenyl-1H-  
 pyrazole-1-carboxamide,

N-[(2-chloro-5-thiazolyl)methyl]-N'-methyl-N''-nitro-guanidine,

N-methyl-N'-(1-methyl-2-propenyl)-1,2-hydrazinedicarbothioamide,

N-methyl-N'-2-propenyl-1,2-hydrazinedicarbothioamide,

O,O-diethyl [2-(dipropylamino)-2-oxoethyl]-ethylphosphoramidothioate.

A mixture with other known active compounds, such as herbicides, or with fertilizers  
 and growth regulators is also possible.

In addition, the compounds of the formula (I) according to the invention also have very good antimycotic actions. They have a very broad spectrum of antimycotic action, in particular against Dermatophytes and yeast fungi, moulds and diphasic fungi (for example against *Candida* species, such as *Candida albicans*, *Candida glabrata*), Epidermophyton species, such as *Epidermophyton floccosum*, *Aspergillus* species, such as *Aspergillus niger* and *Aspergillus fumigatus*, *Trichophyton* species, such as *trichophyton mentagrophytes*, *Microsporon* species, such as *Microsporon canis* and *audouinii*. The list of these microorganisms by no means represents a limitation of the mycotic spectrum that can be controlled, but is only of illustrative character.

The active compounds can be employed as such, in the form of their formulations or the use forms prepared therefrom, such as ready-to-use solutions, suspensions, wettable powders, pastes, soluble powders, dusts and granules. Application is carried out in a customary manner, for example by watering, atomizing, spraying, broadcasting, dusting, foaming, spreading, and the like. It is furthermore possible to apply the active compounds by the ultra-low-volume method, or to inject the preparation of active compound or the active compound itself into the soil. It is also possible to treat the seed of the plants.

When using the active compounds according to the invention as fungicides, the application rates can be varied within a relatively wide range, depending on the type of application. In the treatment of parts of plants, the active compound application rates are generally between 0.1 and 10,000 g/ha, preferably between 10 and 1000 g/ha. In the treatment of seed, the active compound application rates are generally between 0.001 and 50 g per kilogram of seed, preferably between 0.01 and 10 g per kilogram of seed. In the treatment of the soil, the active compound application rates are generally between 0.1 and 10,000 g/ha, preferably between 1 and 5000 g/ha.



**Preparation Examples****Example 1:**

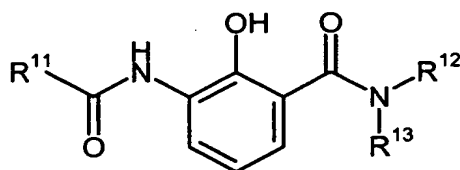
5      Ethyl      2-{{3-(formylamino)-2-hydroxybenzoyl}amino}-3-(4-hydroxyphenyl)-  
propanoate

Process b)

10      2.0 g (5.1 mmol) of ethyl 2-[(2-hydroxy-3-nitrobenzoyl)amino]-3-(4-hydroxy-  
phenyl)propanoate were suspended in 60 ml of formic acid and admixed with 2.0 g  
of Raney nickel. The mixture was stirred at 90°C for 1 hour and then filtered. The  
filtrate was concentrated and the residue was taken up in dichloromethane and  
washed with dist. water. The organic phase was dried over sodium sulphate and then  
15      concentrated to dryness. Purification was carried out on silica gel using the eluent  
mixture ethyl acetate/cyclohexane in a ratio of 6:1. This gives 1.34 g (70% of theory)  
of ethyl 2-{{3-(formylamino)-2-hydroxybenzoyl}amino}-3-(4-hydroxyphenyl)-  
propanoate.

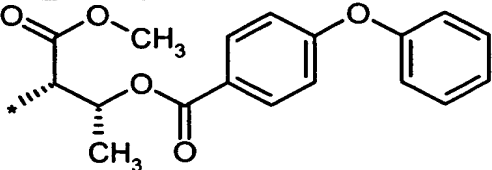
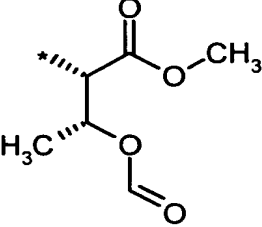
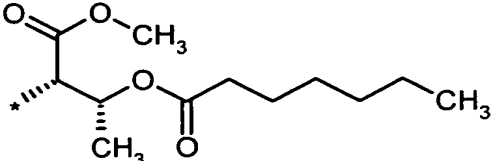
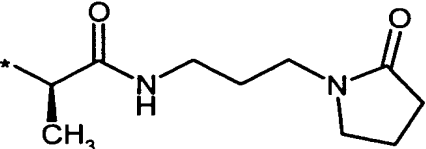
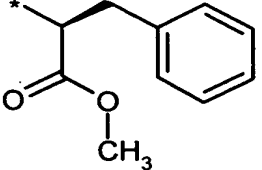
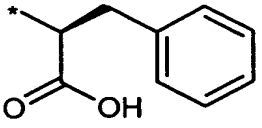
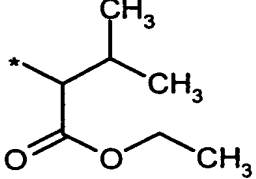
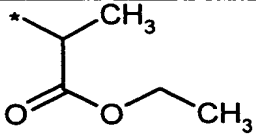
HPLC: logP = 1.83

20      The compounds of the general formula (I-a) listed in Table 1 below are obtained  
analogously to Example 1 and in accordance with the general description of the  
processes according to the invention:

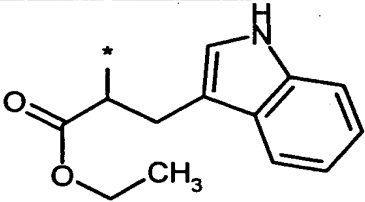
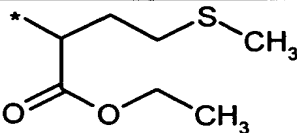
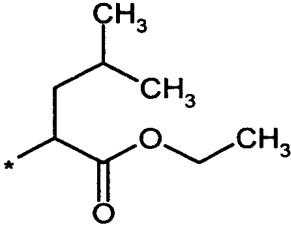
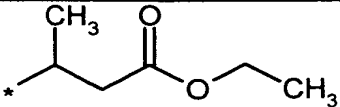
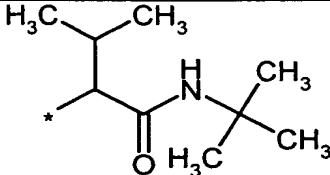
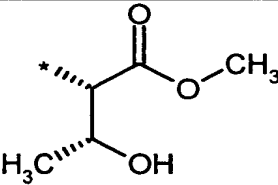
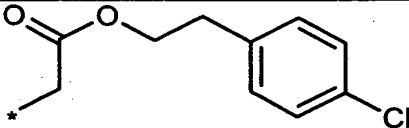
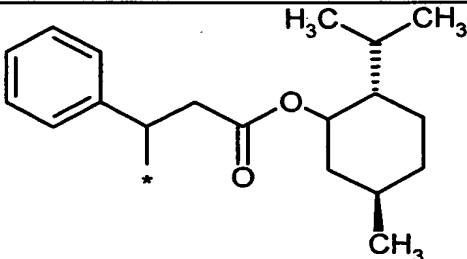


(I-a)

**Table 1:**

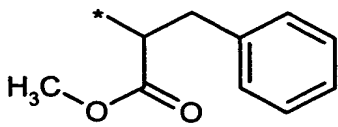
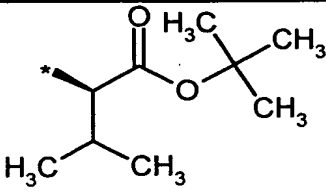
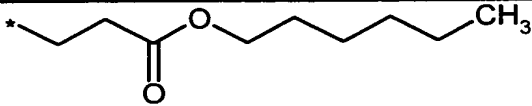
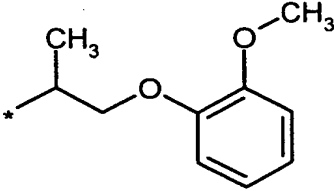
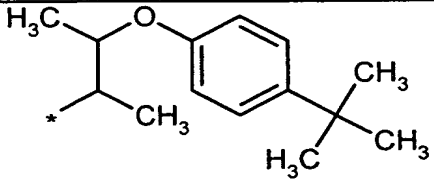
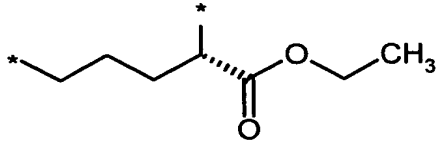
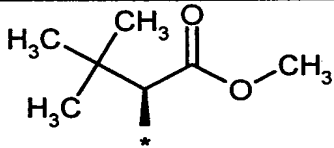
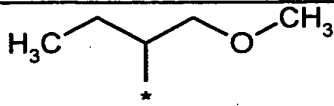
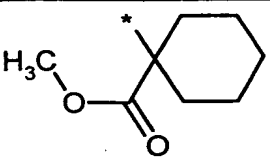
Ex.	R11	R13	R12	logP
2	-H		-H	3.7
3	-H		-H	1.72
4	-H		-H	3.46
5	-CH3		-H	
6	-H		-H	2.21
7	-H		-H	1.73
8	-H		-H	2.26
9	-H		-H	1.64

**Table 1:**

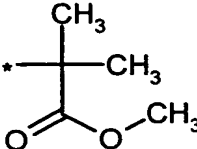
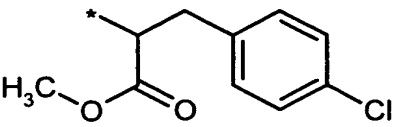
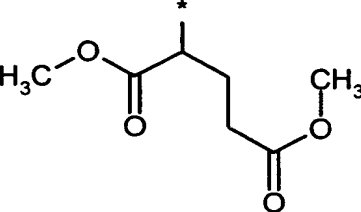
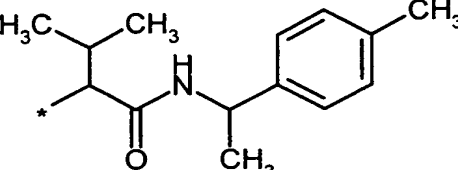
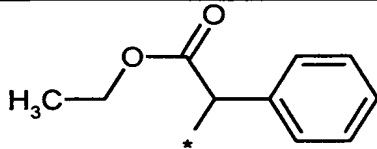
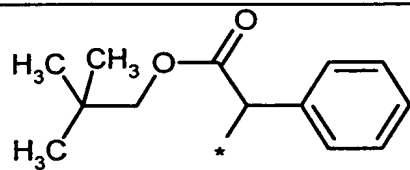
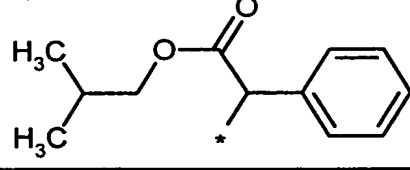
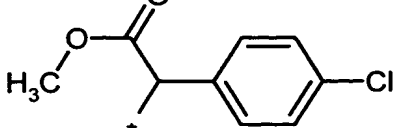
Ex.	R11	R13	R12	logP
10	-H		-H	2.41
11	-H		-H	2.13
12	-H		-H	2.61
13	-H		-H	1.73
14	-H		-H	2.17
15	-H		-H	
16	-H		-H	2.56
17	-H		-H	4.71

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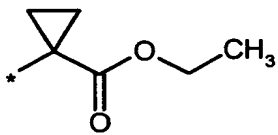
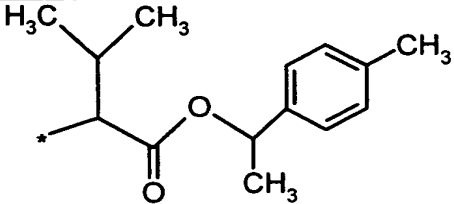
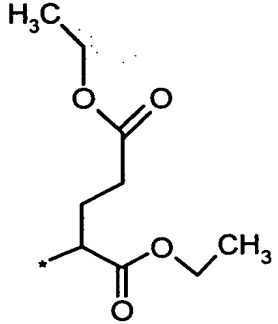
**Table 1:**

Ex.	R11	R13	R12	logP
18	-H		-H	2.27
19	-H		-H	1.52
20	-H		-H	2.93
21	-H		-H	2.36
22	-H		-H	4.03
23	-H			1.55
24	-H		-H	2.29
25	-H		-H	1.71
26	-H		-H	2.26

**Table 1:**

Ex.	R11	R13	R12	logP
27	-H		-H	1.61
28	-H		-H	2.65
29	-H		-H	1.58
30	-H		-H	2.65
31	-H		-H	2.45
32	-H		-H	3.39
33	-H		-H	3.11
34	-H		-H	2.57

**Table 1:**

Ex.	R11	R13	R12	logP
35	-H		-H	1.60
36	H		H	3.58
37	H		H	2.10

\* denotes the site of attachment to the nitrogen atom.

The logP values were determined in accordance with EEC Directive 79/831 Annex V. A8 by HPLC (gradient method, acetonitrile/0.1% aqueous phosphoric acid)

**Preparation of intermediates of the formula (IB)**

**Example (IV-1)**

5 Ethyl 2-[(2-hydroxy-3-nitrobenzoyl)amino]-3-(4-hydroxyphenyl)propanoate.

Process d)

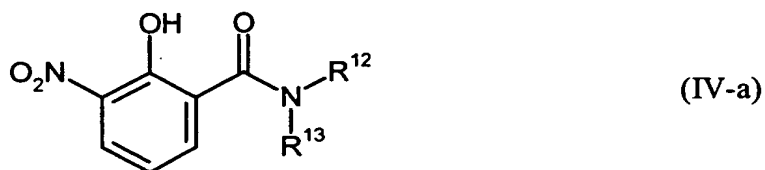
10 1.78 g (8.0 mmol) of D,L-tyrosine ethyl ester hydrochloride are dissolved in 20 ml of tetrahydrofuran and admixed with 1.1 ml (8 mmol) of triethylamine. At 0°C, 1.61 g (8.0 mmol) of 3-nitrosalicylic acid chloride, dissolved in 25 ml of tetrahydrofuran, are added dropwise with stirring to the reaction mixture. Over the course of 16 hours, the reaction mixture is allowed to warm to room temperature. The precipitated triethylammonium chloride is filtered off and the solution that remains is

15 concentrated using a rotary evaporator. The residue is taken up in 200 ml of ethyl acetate and the mixture is extracted with 200 ml of dist. water. The organic phase is subsequently dried over sodium sulphate. The solvent is removed using a rotary evaporator. Purification is carried out over silica gel using the eluent mixture ethyl acetate/cyclohexane in a ratio of 10:1.

20 This gives 2.14 g (69% of theory) of ethyl 2-[(2-hydroxy-3-nitrobenzoyl)amino]-3-(4-hydroxyphenyl)propanoate.

HPLC: logP = 2.28

The compounds of the general formula (IV) listed in Table 2 below are obtained analogously to Example (IV-1) and in accordance with the general description of the processes according to the invention:



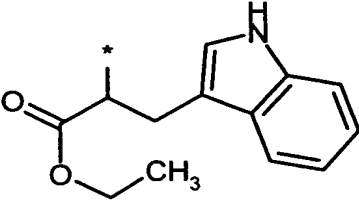
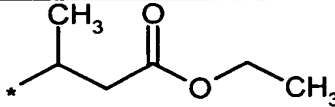
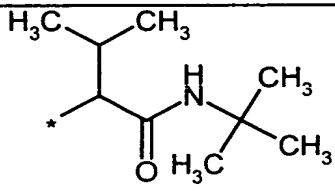
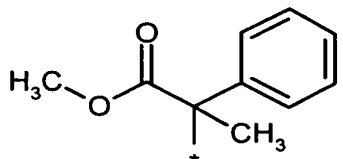
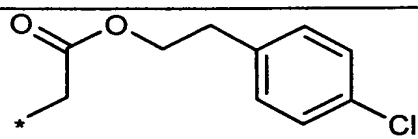
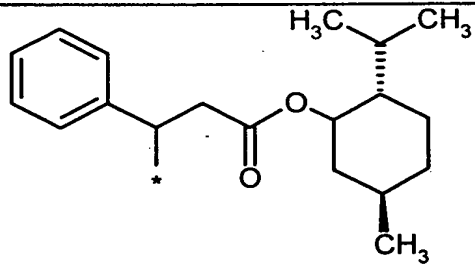
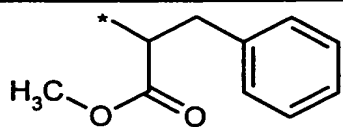
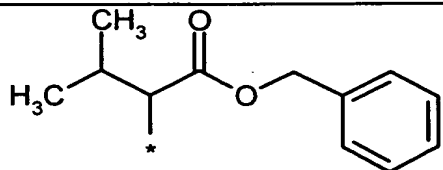
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**Table 2:**

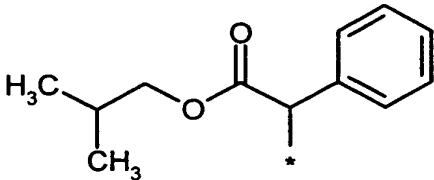
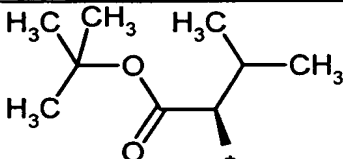
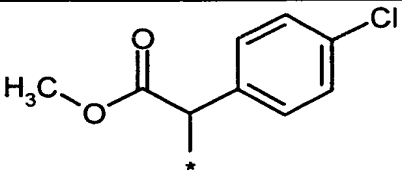
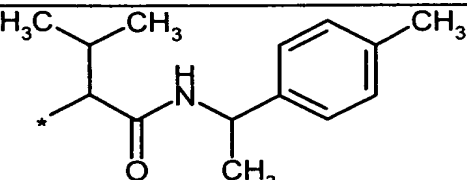
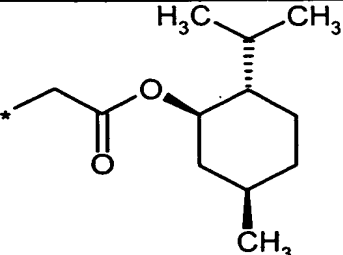
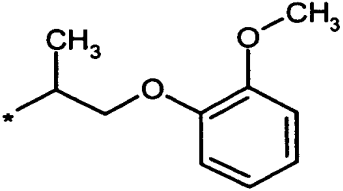
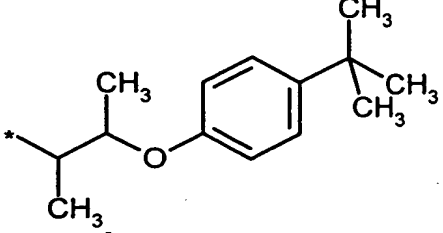
Ex. No.	R <sup>12</sup>	R <sup>13</sup>	LogP
IV-2	-H	-CH <sub>2</sub> -COOCH <sub>3</sub>	1.47
IV-3	-H		2.79
IV-4	-H		2.16
IV-5	-H		2.82
IV-6	-H		2.68
IV-7	-H		3.23



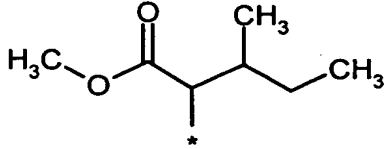
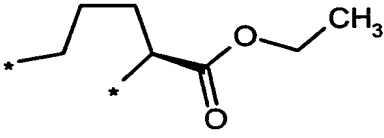
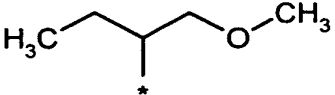
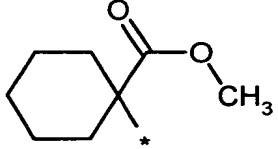
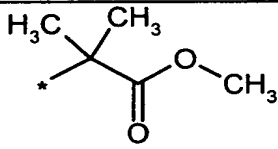
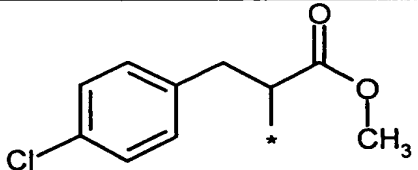
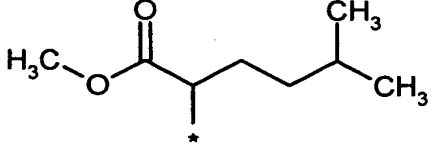
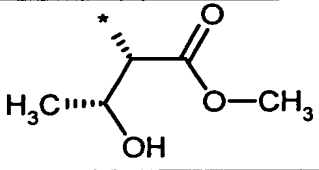
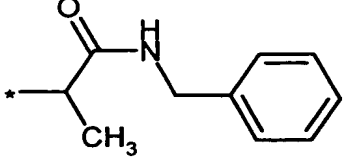
**Table 2:**

Ex. No.	R <sup>12</sup>	R <sup>13</sup>	LogP
IV-8	-H		2.93
IV-9	-H		2.25
IV-10	-H		2.65
IV-11	-H		2.85
IV-12	-H		3.1
IV-13	-H		5.28
IV-14	-H		2.78
IV-15	-H		3.48

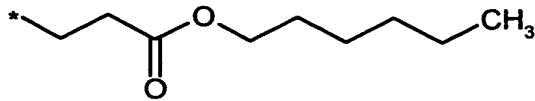
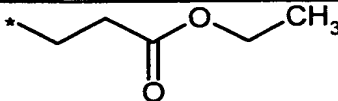
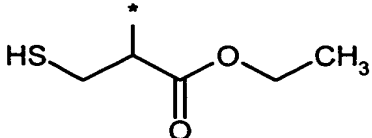
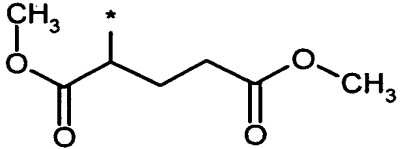
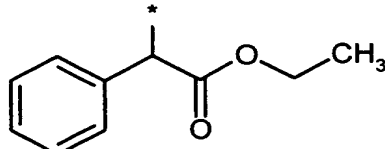
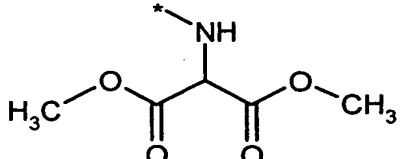
**Table 2:**

Ex. No.	R12	R13	LogP
IV-16	-H		3.99
IV-17	-H		3.53
IV-18	-H		3.1
IV-19	-H		3.09
IV-20	-H		4.4
IV-21	-H		2.91
IV-22	-H		4.63

**Table 2:**

Ex. No.	R12	R13	LogP
IV-23	-H		2.83
IV-24			1.86
IV-25	-H		2.19
IV-26	-H		2.73
IV-27	-H		2.05
IV-28	-H		3.14
IV-29	-H		2.89
IV-30	-H		1.44
IV-31	-H		MPLC: m/e = 390.9

**Table 2:**

Ex. No.	R <sup>12</sup>	R <sup>13</sup>	LogP
IV-32	-H		3.51
IV-33	-H		2.01
IV-34	-H		3.35
IV-35	-H		1.99
IV-36	-H		2.98
IV-37	-H		

\* denotes the site of attachment to the nitrogen atom.

The logP values were determined in accordance with EEC Directive 79/831 Annex V. A8 by HPLC (gradient method, acetonitrile/0.1% aqueous phosphoric acid)

**Use Examples:**

**Example A**

5      **Phytophthora test (tomato) / protective**

Solvent:    24.5 parts by weight of acetone

             24.5 parts by weight of dimethylacetamide

Emulsifier: 1.0 part by weight of alkylaryl polyglycol ether

10

To produce a suitable preparation of active compound, 1 part by weight of active compound is mixed with the stated amounts of solvent and emulsifier, and the concentrate is diluted with water to the desired concentration.

15

To test for protective activity, young plants are sprayed with the preparation of active compound at the stated application rate. After the spray coating has dried on, the plants are inoculated with an aqueous spore suspension of *Phytophthora infestans*. The plants are then placed in an incubation cabin at approximately 20°C and 100% relative atmospheric humidity.

20

Evaluation is carried out 3 days after the inoculation. 0% means an efficacy which corresponds to that of the control, whereas an efficacy of 100% means that no infection is observed.

25

In this test, the substances according to the invention listed in Examples (2), (4), (8) and (12) exhibit, at an application rate of 100 g/ha, an efficacy of 92% or more.

**Example B**

Plasmopara test (grapevine) / protective

- 5        Solvent:    24.5 parts by weight of acetone  
                 24.5 parts by weight of dimethylacetamide  
Emulsifier: 1.0    part by weight of alkylaryl polyglycol ether

10        To produce a suitable preparation of active compound, 1 part by weight of active compound is mixed with the stated amounts of solvent and emulsifier, and the concentrate is diluted with water to the desired concentration.

15        To test for protective activity, young plants are sprayed with the preparation of active compound at the stated application rate. After the spray coating has dried on, the plants are inoculated with an aqueous spore suspension of *Plasmopara viticola* and then remain in an incubation cabin at approximately 20°C and 100 % relative atmospheric humidity for 1 day. The plants are then placed in a greenhouse at approximately 21°C and approximately 90% atmospheric humidity for 5 days. The plants are then moistened and placed in an incubation cabin for 1 day.

20        Evaluation is carried out 6 days after the inoculation. 0% means an efficacy which corresponds to that of the control, whereas an efficacy of 100% means that no infection is observed.

25        In this test, the substances according to the invention listed in Examples (2), (4), (6), (8) and (12) exhibit, at an application rate of 100 g/ha, an efficacy of 97% or more.

**Example C**

Botrytis test (beans) / protective

- 5        Solvent:    24.5 parts by weight of acetone  
                     24.5 parts by weight of dimethylacetamide  
      Emulsifier: 1.0    part by weight of alkylaryl polyglycol ether

10        To produce a suitable preparation of active compound, 1 part by weight of active compound is mixed with the stated amounts of solvent and emulsifier, and the concentrate is diluted with water to the desired concentration.

15        To test for protective activity, young plants are sprayed with the preparation of active compound at the stated application rate. After the spray coating has dried on, 2 small pieces of agar overgrown with Botrytis cinerea are placed onto each leaf. The inoculated plants are placed in a dark chamber at approximately 20°C and 100% relative atmospheric humidity.

20        The size of the infected areas on the leaves is evaluated 2 days after the inoculation. 0% means an efficacy which corresponds to that of the control, whereas an efficacy of 100% means that no infection is observed.

In this test, the substances according to the invention listed in Examples (2), (4), (6), (8) and (12) exhibit, at an application rate of 500 g/ha, an efficacy of 90% or more.

**Example D**

**Pyricularia test (rice) / protective**

- 5        Solvent:    48.8        parts by weight of acetone  
      Emulsifier: 1.2        parts by weight of alkylaryl polyglycol ether

10        To produce a suitable preparation of active compound, 1 part by weight of active compound is mixed with the stated amount of solvent and emulsifier, and the concentrate is diluted with water to the desired concentration.

15        To test for protective activity, young rice plants are sprayed with the preparation of active compound at the stated application rate. After the spray coating has dried on, the plants are inoculated with an aqueous spore suspension of *Pyricularia oryzae* and then remain at 100% rel. atmospheric humidity and 26°C for 24 h. The plants are then placed in a greenhouse at 80% relative atmospheric humidity and a temperature of 26°C.

20        Evaluation is carried out 7 days after the inoculation. 0% means an efficacy which corresponds to that of the control, whereas an efficacy of 100% means that no infection is observed.

In this test, the substances according to the invention listed in Examples (1), (8), (9), (10) and (12) exhibit, at an application rate of 750 g/ha, an efficacy of 89% or more.